



## **Greenhouse gas emissions from wastewater treatment plants: measurements and carbon footprint assessment**

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# Greenhouse gas emissions from wastewater treatment plants: measurements and carbon footprint assessment



Antonio Delre

PhD Thesis  
February 2018





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Department of Environmental Engineering  
Technical University of Denmark

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The synopsis part of this thesis is available as a PDF file for download from the DTU research database ORBIT: <http://www.orbit.dtu.dk>.

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Photos shot by Antonio Delre at Tekniska verken in Linköping, Sweden, (top-left), and at Lynetten wastewater treatment plant (Biofos) in Copenhagen, Denmark, (bottom-right).

To my grandma Giovanna,  
the teacher who has unwittingly supported this adventure

*“e volta nostra poppa nel mattino,  
de’ remi facemmo ali al folle volo”*

*“and having turned our stern unto the morning,  
we of the oars made wings for our mad flight”*

Ulysses in *The Divine Comedy, Inferno, Canto XXVI*  
by Dante Alighieri

Translation by Henry Wadsworth Longfellow

# Preface

The work presented in this PhD thesis was carried out at the Department of Environmental Engineering of the Technical University of Denmark from December 2013 to November 2017. This thesis was prepared as part of the LaGas project (<http://www.lagas.dk>). The research was performed under the main supervision of Professor Charlotte Scheutz (DTU Environment) and co-supervision of Dr Jacob Mønster (Industrial Post-Doc at DTU Environment and FORCE Technology).

The thesis is organised in two parts: the first part puts into context the findings of the PhD in an introductory review, while the second part consists of the papers listed below. These will be referred to in the text by their paper number, written in Roman numerals **I-V**.

- I. Delre A., Mønster J., Samuelsson J., Fredenslund A. M., Scheutz C., 2017. Emission quantification using the tracer gas dispersion method: the influence of instrument, tracer gas species and source simulation. Manuscript submitted to *Science of the Total Environment*, November 2017.
- II. Delre A., Mønster J., Scheutz C., 2017. Greenhouse gas emission quantification from wastewater treatment plants, using a tracer gas dispersion method. *Science of the Total Environment* 605–606, 258–268. doi:10.1016/j.scitotenv.2017.06.177
- III. Samuelsson J., Delre A., Tumlin S., Hadi S., Offerle B., Scheutz C., 2017. Optical technologies applied alongside on-site and remote approaches for climate gas emission quantification at a wastewater treatment plant. Manuscript submitted to *Water Research*, September 2017.
- IV. Reinelt T., Delre A., Westerkamp T., Holmgrend M. A., Liebetrau J., Scheutz C., 2017. Comparative use of different emission measurement approaches to determine methane emissions from a biogas plant. *Waste Management* 68, 173–185. doi:10.1016/j.wasman.2017.05.053



- V. Delre A., ten Hoeve M., Scheutz C., 2017. Site-specific carbon footprints of Scandinavian wastewater treatment plants using the life cycle assessment approach. Manuscript submitted to *Water Research*, November 2017.

In this online version of the thesis, paper **I-V** are not included but can be obtained from electronic article databases e.g. via [www.orbit.dtu.dk](http://www.orbit.dtu.dk) or on request from DTU Environment, Technical University of Denmark, Bygningstorvet, Building 115, 2800 Kgs. Lyngby, Denmark, [info@env.dtu.dk](mailto:info@env.dtu.dk).

In addition, the following publications, not included in this thesis, were also concluded during this PhD study:

- Delre A, Mønster J., Scheutz C. (2014). Full-scale quantification of CH<sub>4</sub> emissions from wastewater treatment plants and biogas facilities. Conference abstract from 2<sup>nd</sup> IBBA Methane emission workshop, Kiel, Germany – September 4<sup>th</sup>, 2014. Abstract available at <http://conference.sgc.se/ckfinder/userfiles/files/DelreDTU.pdf> (last accessed on November 18<sup>th</sup>, 2017)
- Holmgren, M. A., Hansen M. N. Reinelt T., Westerkamp T., Jørgensen L. Scheutz C., Delre A. (2015). Measurements of methane emissions from biogas production. EnergiForsk report 2015:158. ISBN (electronic) 978-91-7673-158-1.



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I would also like to thank *Mr Jerker Samuelsson* (Flux Sense AB) for his important contribution and suggestions during meetings and long joint measurement campaigns. I thank *Mr Magnus Andreas Holmgren* (RISE – Research Institute of Sweden) for welcoming me onto the international project “Measurements of methane emissions from biogas production,” led by Energiforsk AB. All my co-authors are acknowledged for their help and contribution to the papers included in this PhD thesis. I extend my thanks also to *Mr Erik Rønn Lange* for his efficient technical support on the mobile analytical platform I used for my measurements.

These years at DTU Environment, and in general in Copenhagen, have been crucial to my life, and I am really grateful for the number of valuable people I met. Special thanks go to my friends *Bettino* and *Pablito*, precious companions over these years. I warmly thank *Aiga*, important gift in the first part of this adventure. The whole *Solid Waste Research Group* and people at *DTU Environment* are thanked for making these years special. In particular, I wish to thank *Vincent* for being always welcoming, *Alessio* for his open-minded vision and *Ehsan*, *Valentina* and *Teme* for being partners in smiles. *Giorgetta*, *Susanna* and *Concetta* are thanked because they are “scocchiate” – just the way I like. I thank *Fabio* for his smile and support in the last few months of this work. I thank *Sara*, the sweet *Lotte* and *Andreas* for being great office-mates and with whom sharing good and not so good working hours was al-

ways a great experience. I extend my gratitude to *Anders F.* for being a good partner in many measurement campaigns and providing valuable feedbacks to my work. I thank *Nynne* for her simple sweetness and *Gregory* for his stimulating friendship. I also wish to thank *Sanne* for hosting me at her home and being supportive, especially in the last few months of my PhD education.

There are friends that remain with me wherever I may roam, regardless of where in the world I go. This work was therefore achieved thanks to the long-distance support of *Baba*, *il Profeta*, *Cekkella*, *Phil*, *Daba* and *Peppe Fronte-Retro*. I thank them for being the added value in my life.

Last but not least, I wish to thank my family for being my strength, my backbone, my wisdom, my shelter, my stable roots. I thank my beloved mother and father for being the Great Bow from where I was shot free, and I thank my much-loved brother, who enjoys growing up with me.

# Summary

The anthropogenic emissions of greenhouse gases (GHGs) into the atmosphere are of great concern, due to their effect on climate change. To curb the increased accumulation of GHGs in the atmosphere, the United Nations Framework Convention on Climate Change promotes the reporting of national anthropogenic GHG emissions. Wastewater treatment plants (WWTPs) emit two potent GHGs, namely methane ( $\text{CH}_4$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ), but also carbon dioxide ( $\text{CO}_2$ ), which is not accounted as a GHG, due to its biogenic origin (IPCC, 2006). Currently,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions from WWTPs are estimated according to national and international guidelines. However, it is unknown how well these estimated emissions resemble actual plant-specific  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions; therefore,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emission quantifications are needed to assure the reliable accounting of GHG emissions on the plant scale.

Quantifying GHG emissions from WWTPs is a challenging undertaking, since emissions are fugitive, and are occurring across a large area consisting of several smaller sources at different emission heights and physical shapes. In the last two decades, this challenge has been addressed mainly by using on-site point measurements, namely measuring directly on individual emitting sources identified inside the facility. Usually, a single source is either completely encapsulated or partially covered, in order to measure GHG concentration in a known air volume or through a known air flow, i.e. the flux chamber techniques. Complete encapsulation of a single emitting source usually occurs when measuring leakages from biogas bearing process units, while partial coverage usually is used when measuring emissions from surfaces such as open basins surface. In the last few years, plant-integrated measurements have also been performed at WWTPs, using the mobile tracer gas dispersion method (MTDM). This method uses a ground-based remote sensing approach combining the controlled release of tracer gas from the WWTP with atmospheric plume concentration measurements. Since facilities having different plant layouts, and using different process units and technologies could require different applications of the method, further investigations are needed to identify how MTDM can be best applied at WWTPs.

In addition to fugitive emissions of  $\text{CH}_4$  and  $\text{N}_2\text{O}$ , WWTPs indirectly emit GHGs, mainly  $\text{CO}_2$ , due to the consumption of chemicals and energy. The carbon footprint assessment allows the quantification of the overall contribution of a WWTP to climate change.

The principal aim of this PhD thesis was to implement the MTDM application at WWTPs, in order to quantify plant-integrated CH<sub>4</sub> and N<sub>2</sub>O emissions. Additionally, the influence of analytical instrument characteristics and tracer gas release on MTDM results was investigated. GHG emissions were quantified at six WWTPs, using the MTDM. At two facilities, plant-integrated and on-site point measurement approaches were compared. Finally, the PhD thesis assesses the importance of fugitive GHG emissions in the carbon footprint evaluation of WWTPs.

Fair agreement of the plant-integrated CH<sub>4</sub> emission rates was obtained when three analytical instruments, with different detection frequencies and precisions, were used in a simultaneous MTDM application at a WWTP. Emission rates differed between 1 and 18% from the mean emission rate quantified by all instruments. In the same campaign, the importance of a high plume signal within a plume traverse was shown, as this resulted in more reliable plant-integrated emission quantifications.

An estimation of the MTDM detection limit was introduced by using inverse Gaussian plume modelling. Estimating the lowest detectable emission rate by MTDM was useful when little GHG was emitted from a large area, forcing measurements at a distance so far away that the analytical instrument, due to high atmospheric dilution, could not distinguish the plume from background concentrations within the plume traverse.

The introduction of an indicator called “Underestimation due to Tracer Height release” (UTH) was useful in documenting that a potential vertical misplacement of the tracer gas had little influence on the final emission rate quantification. Information provided by UTH is relevant when the plume is traversed relatively close to a facility with emissions occurring from elevated heights, which incurs risk of underestimating the CH<sub>4</sub> emission, because tracer gas released from the ground does not mix completely with CH<sub>4</sub> potentially emitted from the top of a CH<sub>4</sub>-bearing process unit.

The relevance of correct target gas simulation using proper tracer gas placement was demonstrated by applying MTDM for plant-integrated CH<sub>4</sub> quantification. A minor emission rate overestimation was caused by a sideward misplacement of 250 m, whereas the emission rate overestimation increased by up to almost 50% when the tracer gas was misplaced 150 m upwind of the correct position. This large error in emission rate quantification was caused by the different travel distances of the target and the tracer gases.

Plant-integrated CH<sub>4</sub> emission rates were between 1.1 and 39.5 kg CH<sub>4</sub> h<sup>-1</sup>, and corresponding CH<sub>4</sub> emission factors were between 1.1% and 21.3% as kg CH<sub>4</sub> (kg CH<sub>4</sub> production)<sup>-1</sup> and between 0.2% and 3.2% as kg CH<sub>4</sub> (kg COD influent)<sup>-1</sup>. Plant-integrated N<sub>2</sub>O emission rates were between < 0.1 and 6.4 kg N<sub>2</sub>O h<sup>-1</sup>, and corresponding N<sub>2</sub>O emission factors were between < 0.1% and 5.2% as kg N<sub>2</sub>O-N (kg TN influent)<sup>-1</sup>.

A comparison of plant-integrated and on-site measurements at two facilities showed that plant-integrated measurements generally provided more comprehensive emission quantifications – most likely because on-site methods may not quantify all emission sources. Plant-integrated methods are thus useful for carbon footprint evaluations of an entire facility – and thus for emission reporting. On-site approaches provided information about emissions occurring from specific sources identified inside the facility, which is important in the daily operation of the plant in optimising treatment technologies and reducing emissions.

The carbon footprint was assessed at seven WWTPs. None of the utilities could be considered carbon-neutral, due to their positive net carbon footprint. The assessment revealed that fugitive GHG emissions were very important when evaluating the impact of wastewater utilities on climate change, because they could contribute up to 71% of the total burden. The importance of accurate GHG emission factors was highlighted by the performed sensitivity and scenario analysis. GHG emission factors were found to be sensitive model parameters, small changes to which led to large changes in carbon footprint results. GHG emission factors were also largely responsible for the uncertainty of the net carbon footprint evaluation. Additionally, using default values provided in emission reporting guidelines gave a net carbon footprint up to four times smaller or seven times larger compared to when measured plant-integrated GHG emission rates were used. Finally, when the carbon footprint was evaluated by comparing fugitive CH<sub>4</sub> emissions measured during normal operational conditions and digester malfunctioning, the results were up to 320 times higher when problems at digesters occurred, suggesting a careful monitoring strategy that includes emission variations in the inventory year.





# Dansk sammenfatning

Antropogene emissioner af drivhusgasser (GHG) til atmosfæren bør begrænses på grund af deres påvirkning af klimaet. For at bremse ophobningen af drivhusgasser i atmosfæren, anbefaler FN's rammekonvention for klimaændringer at nationale antropogene drivhusgasemissioner indrapporteres. Spildevandsrensningsanlæg udleder metan ( $\text{CH}_4$ ) og lattergas ( $\text{N}_2\text{O}$ ) - to potente drivhusgasser - til atmosfæren. Der udledes også kuldioxid ( $\text{CO}_2$ ), som i denne sammenhæng ikke betragtes som drivhusgas på grund af dets biogene oprindelse (IPCC, 2006). I øjeblikket estimeres  $\text{CH}_4$ - og  $\text{N}_2\text{O}$ -emissioner fra spildevandsrensningsanlæg i henhold til nationale og internationale retningslinjer. Det er imidlertid uvist, hvor godt disse estimerede emissioner ligner faktiske anlægsspecifikke  $\text{CH}_4$ - og  $\text{N}_2\text{O}$ -emissioner. Derfor er der brug for metoder til at måle  $\text{CH}_4$ - og  $\text{N}_2\text{O}$ -emissioner fra spildevandsrensningsanlæg for at sikre pålidelig estimering og indrapportering af drivhusgasemissioner på anlægsskala. Kvantificering af drivhusgasemissioner fra rensningsanlæg på baggrund af målinger er en udfordrende opgave, da emissionen sker fra flere processer fordelt på et stort område. Disse emissioner stammer fra flere mindre kilder med forskellige emissionshøjder og fysiske udformninger. I de sidste to årtier har emissioner til luften fra rensningsanlæg og lignende anlæg været målt ved at benytte punktmålinger, hvor der måles direkte på de identificerede emitterende kilder på anlægget. Normalt bruges fluxkammertechnikker, hvor en kilde enten fuldstændigt indkapsles eller delvist dækkes for at måle GHG-koncentrationen i et kendt luftvolumen eller i en kendt luftstrøm. Fuldstændig indkapsling af en kilde bruges normalt ved måling af lækager fra procesenheder med biogas, hvorimod delvis tildækning bruges ved måling af emissioner fra overflader såsom bassiner. Inden for de seneste år er der udviklet en jordbaseret telemålemetode baseret på brug af sporgas, der kan måle den samlede gasemission fra et anlæg. Metoden, der kaldes sporgasdispersionsmetoden, har primært været brugt til at måle  $\text{CH}_4$  emission fra deponier, men har inden dette ph.d. studie været afprøvet på to spildevandsrenseanlæg. Målemetoden kombinerer en kontrolleret frigivelse af sporgas fra rensningsanlægget med atmosfæriske fane-koncentrationsmålinger. Rensningsanlæg har forskellige layout og benytter forskellige processer og teknologier til vand- og slambehandling. For at kunne måle emissioner fra rensningsanlæg med MTDM kræves der yderligere undersøgelser af målemetoden - herunder krav til udstyr, opsætning af sporgas samt udførsel af målinger for at identificere, hvordan MTDM bedst kan anvendes på anlæg til spildevandsrensning. Udover udslip af  $\text{CH}_4$  og  $\text{N}_2\text{O}$  udledes der indirekte drivhusgasemissioner fra

spildevandsrensning på grund af forbruget af kemikalier og energi. Brugen af CO<sub>2</sub>-fodafttryk gør det muligt at kvantificere det samlede bidrag til klimaændringerne fra et rensningsanlæg.

Hovedformålet med denne ph.d.-afhandling var at udvide applikationen af MTDM til også at kunne anvendes på rensningsanlæg til at kvantificere anlægsintegrerede CH<sub>4</sub>- og N<sub>2</sub>O-emissioner. Derudover blev indflydelsen af analytiske instrumenters karakteristika samt sporgasfrigivelse undersøgt. Emissionerne af CH<sub>4</sub>- og N<sub>2</sub>O blev kvantificeret på seks spildevandsrensningsanlæg ved hjælp af MTDM. På to anlæg blev anlægsintegrerede emissioner sammenlignet med on-site punktmålinger. Endelig vurderedes vigtigheden af drivhusgasemissioner i evalueringen af et rensningsanlægs CO<sub>2</sub>-fodafttryk.

Måling med tre analytiske instrumenter med forskellige detektionsfrekvenser og nøjagtigheder gav sammenlignelige resultater ved samtidig måling af emissioner fra et rensningsanlæg. Emissionen varierede mellem 1 og 18% fra den gennemsnitlige emission kvantificeret af alle instrumenter. I samme kampagne blev betydningen af et højt fanesignal inden for en fanetraversering belyst, hvilket viste at give en mere pålidelig emissionskvantificering.

Et estimat af MTDM-detektionsgrænsen blev introduceret ved at anvende invers Gaussisk fane modellering. Estimeringen af den laveste detekterbare emission ved brug af MTDM er særlig nyttig på anlæg, hvor emissionen er relativt lavt og sker fra et stort område, hvilket medfører at målingerne må udføres relativt langt nedvinds anlægget. Under sådanne forhold kan det grundet en høj atmosfærisk fortynding være svært at skelne fanen fra baggrundskoncentrationen.

Introduktionen af et udtryk for "Underestimering på grund af sporgasfrigivelseshøjden" (UTH) blev brugt til at dokumentere, hvorvidt en potentiel vertikal fejlplacering af sporgasen kan have haft indflydelse på kvantificeringen af emissionen. UTH er relevant for anlæg hvor emissionen kan ske fra højtplacerede punktudslip, og når fanen er traverseret relativt tæt på anlægget. Her vil der kan være en risiko for at underestimere den målte CH<sub>4</sub>-emission, da sporgassen frigivet ved jordoverfladen ikke er fuldstændig opblandet med CH<sub>4</sub>, der potentielt udsendes fra toppen af en CH<sub>4</sub> førende proces enhed.

Relevansen af korrekt sporgasplacering blev demonstreret ved udførsel af CH<sub>4</sub> emissions målinger på et udvalgt rensningsanlæg. En mindre fejlestime-

ring af emissionen blev set ved en fejlplacering af sporgasfrigivelsen på 250 m til den ene side for kilden, mens emissionen fejlestimeredes med op til næsten 50%, når sporgasen blev fejlplaceret 150 m opvinds fra kilden. Denne fejl i emissionsestimeringen skyldtes, de forskellige afstande imellem kilde og målevej for hhv. kvantificeringsgas ( $\text{CH}_4$ ) og sporgas.

Anlægsintegrerede  $\text{CH}_4$ -emissioner blev målt til mellem 1,1 og 39,5 kg  $\text{CH}_4$   $\text{time}^{-1}$ , og  $\text{CH}_4$  emissionsfaktorer var mellem 1,1% og 21,3% givet som kg  $\text{CH}_4$  (kg produceret  $\text{CH}_4$ ) $^{-1}$  og mellem 0,2% og 3,2% som kg  $\text{CH}_4$  (kg COD influent) $^{-1}$ . Anlægsintegrerede  $\text{N}_2\text{O}$ -emissioner blev målt til mellem <0,1 og 6,4 kg  $\text{N}_2\text{O}$   $\text{time}^{-1}$ , og tilsvarende  $\text{N}_2\text{O}$ -emisisionsfaktorer var mellem <0,1% og 5,2% som kg  $\text{N}_2\text{O-N}$  (kg TN influent) $^{-1}$ .

En sammenligning af anlægsintegrerede emissioner og emissioner målt ved punktmålinger på anlægget viste, at anlægsintegrerede målinger generelt gav højere emissioner. Årsagen til dette er sandsynligvis, at punktmålinger ikke nødvendigvis kvantificerer alle kilder til metanemission. Anlægsintegrerede målinger kan derfor bruges til at give en bedre vurdering af det samlede  $\text{CO}_2$ -fodaft tryk fra et anlæg - og dermed også for indberetningen af emissioner. Direkte målinger på specifikke punktkilder identificeret på anlægget er vigtige i den daglige drift af anlægget for at optimere processerne og reducere emissionerne.

$\text{CO}_2$ -fodaft trykket fra syv rensningsanlæg blev beregnet. Ingen af anlæggene kunne betragtes som værende  $\text{CO}_2$ -neutrale, da de alle havde et positivt netto  $\text{CO}_2$ -fodaft tryk. Vurderingen viste, at drivhusgasemissioner var meget vigtige, i den samlede vurdering af spildevandsanlægs påvirkning af klimaet, da de kunne bidrage med op til 71% af den samlede påvirkning. Betydningen af nøjagtige GHG-emissionsfaktorer blev tydeliggjort af den udførte følsomheds- og scenarieanalyse. GHG-emissionsfaktorerne viste sig at være følsomme modelparametre, hvor små ændringer medførte store ændringer i  $\text{CO}_2$ -fodaft trykket. GHG-emissionsfaktorerne var også i stor udstrækning ansvarlig for usikkerheden på evalueringen af netto  $\text{CO}_2$ -fodaft tryk. Derudover gav brugen af standard-emissionsfaktorer, angivet i vejledninger for emissionsrapportering, et netto  $\text{CO}_2$ -fodaft tryk op til fire gange mindre eller syv gange større sammenlignet med målte anlægsintegrerede emissioner. Endelig, viste  $\text{CO}_2$ -fodaft trykket at være op til 320 gange højere, når emissionsfaktorer målt under driftsproblemer (fejl på rådnetanke) blev anvendt. Dette viser et behov for udarbejdelse af målestrategier, der inkluderer disse emissionsvariationer,

således at der opnås repræsentative emissionsrater og –faktorer, der kan anvendes i den årlige emissionsindrapportering.

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# Abbreviations

CH <sub>4</sub>	Methane
CO <sub>2</sub>	Carbon dioxide
COD	Chemical oxygen demand
DNG	Danish national guidelines
EF	Emission factor
GHG	Greenhouse gas
IDMM	Inverse dispersion modelling method
IPCC	Intergovernmental Panel on Climate Change
LCA	Life cycle assessment
MTDM	Mobile tracer gas dispersion method
N <sub>2</sub> O	Nitrous oxide
NH <sub>3</sub>	Ammonia
OSPM	On-site point measurements
RPR	Release precision ratio
SEM	Standard error of the mean
STDM	Static tracer gas dispersion method
UTH	Underestimation due to tracer height release
WWTP	Wastewater treatment plant





# 1 Introduction

## 1.1 Background and motivations

Anthropogenic activities have changed and continue to change the composition of the Earth's atmosphere. The accumulation of greenhouse gases (GHGs) in the atmosphere modifies the energy balance of the Earth and leads to climate change (IPCC, 2013). The atmospheric concentrations of GHGs have increased from 1750 (preindustrial time) to 2011 as follows: carbon dioxide ( $\text{CO}_2$ ) has increased from 278 to 391 ppm, methane ( $\text{CH}_4$ ) from 722 to 1803 ppm and nitrous oxide ( $\text{N}_2\text{O}$ ) from 259 to 324 ppm (IPCC, 2013). The modification in energy balance of the planet for 2011 relative to 1750 is quantified as radiative forcing, expressed in watts per square metre ( $\text{W m}^{-2}$ ). Positive radiative forcing leads to warming, whereas negative radiative forcing leads to cooling. The radiative forcing of anthropogenic GHG emissions is  $3 \text{ W m}^{-2}$ , and it is composed as follows: 56% is caused by  $\text{CO}_2$ , 32% by  $\text{CH}_4$ , 6% by  $\text{N}_2\text{O}$  and the remaining 6% by halocarbons (IPCC, 2013). Although  $\text{CO}_2$  contributes the most to the radiative forcing of anthropogenic GHG emissions, the emission of the other compounds is also of great concern, due to their high global warming potential, which integrates the radiative forcing over a defined time horizon and provides a value relative to  $\text{CO}_2$  that is the reference gas. The global warming potential of  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  and halocarbons such as trichlorofluoromethane (CFC-11) are, respectively, 28, 265 and 4660, if a time horizon of 100 years is considered (IPCC, 2013). As climate change involves several risks to human and natural systems (IPCC, 2014), a reduction in – or at least stabilisation in the atmosphere of – GHG concentrations is urgent.

To develop solutions for curbing the increasing accumulation of GHGs in the atmosphere, the United Nations Framework Convention on Climate Change encourages the reporting of national anthropogenic GHG emissions. Part of these emissions are generated by wastewater treatment plants (WWTPs), which emit  $\text{CH}_4$  and  $\text{N}_2\text{O}$ , due to biochemical reactions during wastewater and sewage sludge treatment (Daelman et al., 2012; Kampschreur et al., 2009). Wastewater treatment also generates  $\text{CO}_2$  emissions, which, however, are not treated as GHGs due to their biogenic origin (IPCC, 2006). WWTPs operating in the European Union have to report their environmental emissions to an international inventory called the European Pollutant Release and Transfer Register. Although only facilities exceeding specific limits have to

state their emissions, some low-emission plants report their GHG emissions on a voluntary basis (DANVA, 2012; SVU, 2014; Petersson, 2012). Currently, plant operators estimate and report emissions based on simple models provided in national and international guidelines (IPCC, 2006; Thomsen, 2016). However, it is unknown how well these estimates resemble actual plant-specific CH<sub>4</sub> and N<sub>2</sub>O emissions. In this context, site-specific CH<sub>4</sub> and N<sub>2</sub>O emission quantifications are needed, in order to ensure reliable accounting of GHG emissions on the plant scale.

Quantification of GHG emissions from WWTPs are challenging, because CH<sub>4</sub> and N<sub>2</sub>O are emitted from a large area consisting of several smaller sources with different emission heights and physical shapes, resulting in fugitive and diffusive emission patterns. GHG emissions from WWTPs are fugitive, as they are unintended discharges escaping from process units, and they are diffusive because they occur from sources scattered throughout the facility. An additional challenge in GHG emission quantification from WWTPs comes in the form of operational conditions that involve different emissions over time.

In the last two decades, GHG emissions from WWTPs have been quantified by applying on-site point measurements, e.g. flux chamber techniques or similar methods (Czepiel et al., 1993; Masuda et al., 2015; Petersson et al., 2012). The on-site point measurement approach is based on measurements performed directly on one of the emitting sources identified inside the facility. Usually, the source is either completely encapsulated or partially covered, in order to measure GHG concentration in a known air volume or through a known air flux. Complete encapsulation is used often when measuring emissions from a leakage on a process unit bearing CH<sub>4</sub>. Differently, the partial coverage of the emitting source occurs usually when a floating chamber gathers emissions from part of an open basin containing wastewater or sewage sludge.

Recently, GHG emissions from WWTPs have also been quantified using plant-integrated measurements. Yoshida et al. (2014b) and Yver Kwok et al. (2015) applied the mobile tracer gas dispersion method (MTDM) for quantifications of plant-integrated GHG emissions from a WWTP. The MTDM is a ground-based remote sensing approach that combines the controlled release of tracer gas from the WWTP with downwind atmospheric concentration measurements, and lately it has been validated with a known release test and largely applied to CH<sub>4</sub> emission quantifications for landfills (Mønster et al.,

2014; 2015). Although CH<sub>4</sub> emissions from landfills are also fugitive and diffusive, they occur from area sources very different from and much larger than WWTPs; in addition, they use different treatment technologies, resulting in different emission patterns. Landfills are used for solid waste disposal and mainly emit CH<sub>4</sub> from leakages in the landfill cover, and landfill gas and leachate installations. Alternatively, WWTPs treat wastewater and emit both CH<sub>4</sub> and N<sub>2</sub>O from numerous process units involved in wastewater cleaning and sewage sludge treatment. The current application of the MTDM is limited only to two WWTPs. Since facilities with different plant layouts, and using different process units and technologies, could require different ways of applying the method, further investigations are needed to identify how the MTDM can be best applied at WWTPs.

Although the MTDM has been previously validated, only a specific analytical instrument was used. The literature still lacks of a study investigating the influence of different instrument characteristics, such as detection frequency and precision, on associated results.

On-site and remote sensing approaches have been applied at the same facility only by Yver Kwok et al. (2015) for CH<sub>4</sub> emission quantification, in which a direct comparison of total-plant emissions was not possible, because on-site measurements were performed only at process units made by open basins treating wastewater and not applied at the other numerous process units, which, for example, treated sewage sludge. Such measurements resulted in a study reporting plant-integrated emissions measured with a remote sensing approach and emissions from only a few process units measured using an on-site approach. Therefore, the literature is still missing a direct comparison of on-site and remote sensing approaches when measuring total-plant emissions from the same facility.

Fugitive GHG emissions from WWTPs need to be contextualised to identify their importance at the facility level. Operations at a WWTP involve direct and indirect GHG emissions. Direct GHG emissions are fugitives discharged directly into the atmosphere, whereas indirect GHG emissions are caused by the consumption of chemicals and energy (Yoshida et al., 2014a). The overall contribution of a WWTP to climate change can be evaluated by assessing its carbon footprint, a plant environmental indicator which is lately gaining more attention at international, European and national levels (IWA, 2017; EEA, 2014; DANVA, 2012; SVU, 2014). The literature reports only two studies accounting for the annual contribution of a WWTP to climate change. Gustavsson and Tumlin (2013) assessed the carbon footprint of 16 WWTPs,

without using plant-specific data, e.g. plant-specific fugitive GHG emissions, and by adopting an inconsistent accounting method. Conversely, Yoshida et al. (2014a) assessed the carbon footprint of only one WWTP, using plant-specific data and a consistent accounting method. Both studies reported the importance of fugitive GHG emissions when assessing the carbon footprint of a wastewater facility. However, the literature is still lacking a study investigating the importance of fugitive GHG emissions when plant-specific data are used for assessing the carbon footprint of several WWTPs that use different treatment stages and technologies.

## 1.2 Research objectives

The main objective of this PhD thesis was to implement and apply the mobile tracer gas dispersion method (MTDM) for plant-integrated greenhouse gas (GHG) emission quantifications from wastewater treatment plants (WWTPs).

Additionally, this PhD thesis aimed to:

- Investigate the influence of the analytical instrument characteristics and tracer gas release on emission rates obtained by applying the MTDM (Paper I);
- Quantify fugitive CH<sub>4</sub> and N<sub>2</sub>O emissions from WWTPs (Papers II and III);
- Compare the MTDM with other methods, to investigate when to apply plant-integrated and on-site measurement approaches (Paper IV);
- Assess the importance of fugitive GHG emissions in the carbon footprint evaluation of WWTPs (Paper V).

## 2 The emission of greenhouse gases from wastewater treatment plants

This chapter presents the state of the art of GHG emission quantifications from WWTPs and the carbon footprint evaluation of wastewater utilities. The topic is introduced through a section describing process units at a conventional WWTP in South Scandinavia and the potential on-site sources of fugitive CH<sub>4</sub> and N<sub>2</sub>O emissions.

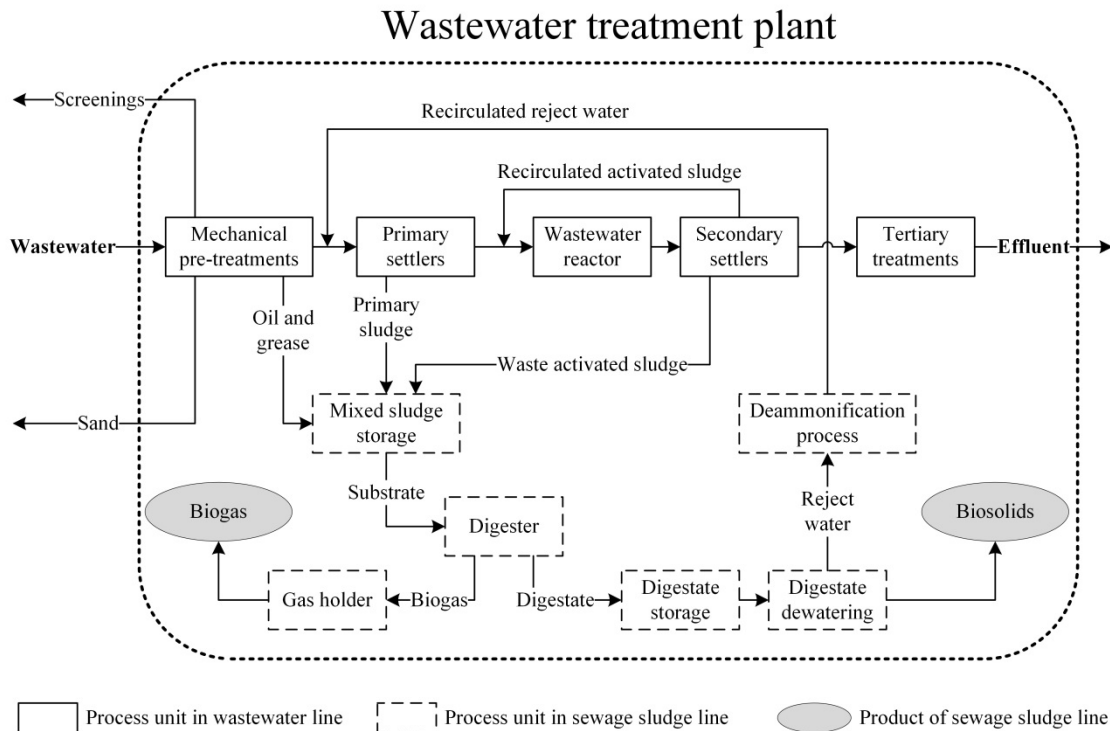
### 2.1 Conventional structure of a wastewater treatment plant

The principal aim of a WWTP is to remove pollution from wastewater and release clean effluent into the environment. Pollution in wastewater is made mainly by organic matter, nutrients such as nitrogen and phosphorus, metals, pathogenic microorganisms and potentially toxic compounds (Metcalf and Eddy, 2004). Wastewater undergoes several treatment stages at the WWTP before clean effluent can be released into the environment.

Figure 1 shows a schematic diagram of a conventional WWTP along with the most common treatment stages in the wastewater and sewage sludge lines. As soon as wastewater enters the WWTP, mechanical pre-treatments remove coarse material (screenings), sand, oil and grease. Screenings are usually burnt in an incineration plant outside the WWTP. Sand is either landfilled or recycled as a filling material in construction works, while oil and grease are sent to the sewage sludge line. After mechanical pre-treatments, primary settlers remove part of the pollution through sedimentation and produce primary sludge, which is sent to the sewage sludge line. In the wastewater reactor, biochemical and chemical reactions separate the remaining pollutants from the wastewater, producing activated sludge. In secondary settlers, the activated sludge is removed from the wastewater and partly recirculated back to the wastewater reactor, in order to improve biochemical reactions. The surplus activated sludge, called “waste activated sludge,” is sent to the sewage sludge line. Before being released into the environment, treated wastewater can undergo a tertiary treatment, which is usually a filtration process removing suspended solids.

In the sewage sludge line, oil and grease, primary sludge and waste activated sludge are mixed and stored before being stabilised in the digester. This material, fed to the digester, is named “substrate.” Anaerobic digestion has two

outputs: biogas and digestate. Biogas is composed mainly of  $\text{CH}_4$  (about 60%) and  $\text{CO}_2$ , and it is stored inside a gasholder before further use, which can vary from plant to plant. In South Scandinavia, biogas is either directly fed into the natural gas grid or upgraded on-site to reach about 99% of  $\text{CH}_4$  content and used as vehicle fuel. An alternative use for biogas in South Scandinavia is by combustion in a combined heat and power engine for the production of heat and electricity. The digestate is stored and subsequently dewatered to produce reject water and biosolids. Reject water is a small flow in comparison to the flow treated in the wastewater line, but it has a higher ammonium content, which could affect microbial activity in the wastewater reactor. For this reason, some plants use a deammonification process to treat reject water before recirculation. Produced biosolids can have different fates, depending on the WWTP: they could be incinerated or stored in stockpiles before being used on agricultural land, due to their nitrogen and phosphorus content.



**Figure 1.** Schematic diagram of a conventional wastewater treatment plant in South Scandinavia. Biogas can be either fed directly into the natural gas grid or used as vehicle fuel after upgrading. Alternatively, biogas can be used for heat and electricity production when burnt in a combined heat and power engine. Biosolids can be either incinerated or stored in stockpiles before being used on agricultural land.

## 2.2 On-site sources of fugitive greenhouse gases

Biological activities during wastewater and sewage sludge treatment involve GHG formation and subsequent fugitive emissions.  $\text{CH}_4$  is produced during the anaerobic degradation of organic matter, due to microbial activity (Metcalf and Eddy, 2004). Since  $\text{CH}_4$  formation already occurs in the sewage network (Liu et al., 2015),  $\text{CH}_4$  can be stripped away during mechanical pre-treatments (Wang et al., 2011; Yan et al., 2014).  $\text{CH}_4$  emitted at the wastewater reactor could originate from an upstream source, such as inlet wastewater and recirculated reject water (Daelman et al., 2012), but it could also be formed in the inner floc region of the activated sludge (Aboobakar et al., 2013b).  $\text{CH}_4$  generated by anaerobic digestion escapes through leakages and the occasional venting of the digester (Liebetrau et al., 2013). However,  $\text{CH}_4$  can also be released before and after digestion, as well as during substrate and digestate storage and/or treatment (Oshita et al., 2014). When biogas is burnt into a combined heat and power engine,  $\text{CH}_4$  could be emitted due to incomplete combustion. Increasing  $\text{CH}_4$  concentration in the biogas via upgrading processes can also cause  $\text{CH}_4$  emissions (Petersson, 2012), while Majumder et al. (2014) reported that biosolids stockpiles emit both  $\text{CH}_4$  and  $\text{N}_2\text{O}$ .

In WWTPs, the microbial activity involved in the nitrification and denitrification processes is used to remove dissolved reactive nitrogen pollution, but it also generates  $\text{N}_2\text{O}$  (Law et al., 2012). Nitrification is an aerobic bacterial respiratory process that releases  $\text{N}_2\text{O}$  as by-product (Inamori et al., 2008), while denitrification is a microbial-mediated process carried out mainly in anoxic conditions, where  $\text{N}_2\text{O}$  is an intermediate step (Tallec et al., 2008). Previous studies have reported  $\text{N}_2\text{O}$  emissions from units performing nitrification and denitrification processes (Kampschreur et al., 2009), often observing  $\text{N}_2\text{O}$  emissions during aeration in the wastewater reactor (Foley et al., 2010; Aboobakar et al., 2013a). The simultaneous study of  $\text{N}_2\text{O}$  in the liquid phase and off-gas suggest that  $\text{N}_2\text{O}$  might be stripped away during aeration, but it can be generated in a different reactor stage (Daelman et al., 2013; Ren et al., 2013). Aboobakar et al. (2013a, 2013b) and Yan et al. (2014) observed large temporal and spatial variability in both  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions across the surface of a wastewater reactor. Finally, the deammonification process, used for reject water treatment, has also been found to be a source of fugitive  $\text{N}_2\text{O}$  emissions (Christensson et al., 2013).



## 2.3 Measurements of fugitive greenhouse gas emissions

The first studies reporting GHG emissions from WWTPs date back to the early 1990s and used on-site point measurements to quantify  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions from open basins in the wastewater treatment line (Czepiel et al., 1993; 1995; Sümer et al., 1995; Wicht and Beier, 1995). More recently, the same process units have been investigated, using a combination of the floating chamber technique and a simultaneous liquid-phase analysis to investigate triggering factors as well as spatial and temporal variations in GHG emissions (Kampschreur et al., 2009; Ren et al., 2013; Rodriguez-Caballero et al., 2014). Long-term investigations have been performed for enclosed treatment technologies with air collection systems to study daily and seasonal variations in GHG emissions (Daelman et al., 2013; Toyoda et al., 2011). However, open-air technologies cannot be investigated with the previous approach, which additionally does not include losses from leaking pipes, fitting and incidental pressure release valves.

Biogas plants usually use manure, organic household waste and organic industrial waste as substrate for  $\text{CH}_4$  production. Biogas plants are equipped with the same technologies and processes used for sewage sludge treatment at WWTPs.  $\text{CH}_4$  emission quantifications from biogas plants have also been carried out by applying on-site point measurements. These investigations usually start with leakage identification and follow with the flux chamber technique for emission measurement (Liebetrau et al., 2013).

Only very recently, GHG emissions from WWTPs and biogas plants have been quantified, using ground-based remote sensing approaches. Some studies have measured  $\text{CH}_4$  emissions from biogas plants, by adopting open-path gas analysers coupled with backward Lagrangian Stochastic modelling, i.e. the inverse dispersion modelling method (IDMM) (Flesch et al., 2011; Groth et al., 2015). To date, only two studies have quantified GHG emissions from WWTPs, using the MTDM (Yoshida et al., 2014b; Yver Kwok et al., 2015).

GHG emissions from different facilities, or emissions recorded at different times at the same facility, can be compared to each other when they are ex-

pressed as emission factors (EFs). In the literature, CH<sub>4</sub> emissions from wastewater treatment reactors are usually normalised by the carbon load going into the WWTP (e.g. kg CH<sub>4</sub> emitted per kg COD<sup>1</sup> influent into the WWTP). Conversely, CH<sub>4</sub> emissions from plants producing biogas are normalised by CH<sub>4</sub> production (e.g. kg CH<sub>4</sub> emitted per kg CH<sub>4</sub> produced). When studying WWTPs, the use of these EFs allows the investigation of CH<sub>4</sub> losses from both treatment capacity and energy points of view. N<sub>2</sub>O emissions are usually normalised by the total nitrogen (TN) influent going into the WWTP (e.g. kg N<sub>2</sub>O-N emitted per kg TN influent), or sometimes by TN removed by the treatment (e.g. kg N<sub>2</sub>O-N emitted per kg TN removed). The former relates N<sub>2</sub>O emissions to the pollution load, while the latter relates N<sub>2</sub>O emissions to the pollution removal capabilities of the treatment.

Table 1 compiles EFs measured at WWTPs, expressed in percentage of kg CH<sub>4</sub> (COD influent)<sup>-1</sup> reported in the literature. CH<sub>4</sub> EFs were between 0.016% and 9.08%. Generally, comparable CH<sub>4</sub> EFs were reported for wastewater reactors and sludge treatment units, while higher CH<sub>4</sub> EFs were reported when whole plants were measured. The highest CH<sub>4</sub> EF was reported by Yoshida et al. (2014b) when measurements were performed during digester malfunctioning. Overall, Table 1 shows that both the wastewater and sludge treatment lines contribute CH<sub>4</sub> emissions from the WWTP.

Table 2 compiles EFs measured at WWTPs, expressed in percentage of kg CH<sub>4</sub> (kg CH<sub>4</sub> produced)<sup>-1</sup> reported in the literature, including EFs used in national and international guidelines (Clean Development Mechanism (CDM) (2012) and Thomsen (2016)). Besides CH<sub>4</sub> EFs measured at WWTPs, Table 2 also reports EFs measured at biogas plants, due to the very few investigations performed at WWTPs and the very similar process units used in the two kinds of facilities. The CH<sub>4</sub> EFs ranged from 0.15% to 32.7% at WWTPs. The lowest value was measured at WWTPs, where only sludge treatment units were investigated (Petersson, 2012), while the largest CH<sub>4</sub> EF was recorded for the whole plant when digesters were malfunctioning (Yoshida et al., 2014b).

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<sup>1</sup> At WWTPs, carbon in wastewater is usually expressed as chemical oxygen demand (COD), which is a measure of the oxygen necessary to oxidise chemically organic material in wastewater when dichromate in an acid solution is used (Metcalf and Eddy, 2004). COD is usually reported as the mass of oxygen consumed per volume of wastewater (e.g. mg l<sup>-1</sup>).

When EFs referring to the malfunctioning of the digesters are neglected, measurements performed only at the sludge treatment units (Petersson, 2012) provided a range of CH<sub>4</sub> EFs lower than measurements for the whole plant (Yoshida et al., 2014b). CH<sub>4</sub> EFs suggested in national and international guidelines cover a large range, from 1.3 to 10% of produced CH<sub>4</sub> (CDM (2012) and Thomsen (2016) in Table 2). The CDM guideline suggests different CH<sub>4</sub> EFs (2.8, 5 and 10% of produced CH<sub>4</sub>), depending on the type of anaerobic digesters used for biogas production when generic organic material is used as substrate (e.g. manure, food waste, sewage sludge, etc.). Thomsen (2016) describes the Danish national guideline for direct GHG emission estimations from WWTPs. For CH<sub>4</sub> emissions occurring at digesters in WWTPs, Thomsen (2016) suggests a CH<sub>4</sub> EF equal to 1.3% of produced CH<sub>4</sub> (Table 2). This EF is based on a study investigating digesters at biogas plants, and it results in lower values than suggested by CDM (2012). CH<sub>4</sub> EFs measured for whole biogas plants are between 0.3 and 26.6% of produced CH<sub>4</sub>, whereby the lowest value is based on on-site point measurements (OSPM), while the highest value is based on ground-based remote sensing measurements when the digesters malfunction (Petersson et al. (2012) and Flesch et al. (2011) in Table 2, respectively). The large range of CH<sub>4</sub> EFs referring to a whole biogas plant is reported by both IDMM and OSPM methods (Table 2).

**Table 1.** Literature review of CH<sub>4</sub> emission factors (EFs) measured at wastewater treatment plants, expressed in percentage as kg CH<sub>4</sub> (kg COD influent)<sup>-1</sup>

Source	Method	Value (min – max)	Study
Reactors	OSPM	0.29	Czepiel et al. (1993)
Reactors	OSPM	0.53 – 1.2	STOWA (2010)
Reactors	OSPM	0.034	Toyoda et al. (2010)
Reactors	OSPM	0.06 – 0.1	Wang et al. (2011)
Reactors	OSPM	0.04 – 0.1	Aboobakar et al. (2013b)
Reactors	OSPM	0.25	Gustavsson and Tumlin (2013)
Reactors	OSPM	0.046 – 1.33	Ren et al. (2013)
Reactors	OSPM	0.016	Rodriguez-Caballero et al. (2014)
Reactors	OSPM	0.15 – 0.69	Masuda et al. (2015)
Sludge treatment units	OSPM	0.16 – 0.6	Oshita et al. (2014)
Whole plant	OSPM	1.13	Daelman et al. (2012)
Whole plant	MTDM	0.61 – 4.35	Yoshida et al. (2014b)
Whole plant	MTDM	9.08 <sup>a</sup>	Yoshida et al. (2014b)

Values are reported either as single or as interval. <sup>a</sup> Value measured during digester malfunctioning. Reactors refer to the process unit in Figure 1, named “Wastewater reactor”. OSPM: on-site point measurements. MTDM: Mobile tracer gas dispersion method

**Table 2.** Literature review of CH<sub>4</sub> emission factors (EFs) expressed in percentage as kg CH<sub>4</sub> (kg CH<sub>4</sub> production)<sup>-1</sup>

Facility	Source	Method	Value (min – max)	Study
WWTP	Whole plant	MTDM	2.1 – 4.4	Yoshida et al. (2014b)
WWTP	Whole plant	MTDM	32.7 <sup>a</sup>	Yoshida et al. (2014b)
WWTP	Sludge treatment units	OSPM	0.15 – 2.6	Petersson (2012)
Biogas plant	Digester	NA	2.8	CDM (2012)
Biogas plant	Digester	NA	5	CDM (2012)
Biogas plant	Digester	NA	10	CDM (2012)
Biogas plant	Digester	OSPM	1.3	Thomsen (2016)
Biogas plant	Whole plant	IDMM	0.8 – 3.2	Flesch et al. (2011)
Biogas plant	Whole plant	IDMM	26.6 <sup>a</sup>	Flesch et al. (2011)
Biogas plant	Whole plant	IDMM	4	Groth et al. (2015)
Biogas plant	Whole plant	OSPM	0.3 – 5.25	Petersson (2012)
Biogas plant	Whole plant	OSPM	1.1 – 13.7	Liebetrau et al. (2013)

Values are reported either as single or as interval. WWTP: Wastewater treatment plant. CDM: Clean development mechanism EFs according to the type of digester. Thomsen (2016) describes the Danish national guidelines for direct GHG emission estimation from WWTPs. OSPM: on-site point measurements. MTDM: Mobile tracer gas dispersion method. IDMM: Inverse dispersion modelling method. NA: Not available.

<sup>a</sup> Values measured during digester malfunctioning.

Table 3 reports literature-based N<sub>2</sub>O EFs expressed in percentage as kg N<sub>2</sub>O-N (kg TN influent)<sup>-1</sup>. The majority of the studies refer to emissions from wastewater reactors and report a very large range, from 0.001 to 5.7% as kg N<sub>2</sub>O-N (TN influent)<sup>-1</sup>. Studies reporting N<sub>2</sub>O EFs referring to more than one process unit in the wastewater treatment line equally provide a very large range, from 0.014 to 9.3% as kg N<sub>2</sub>O-N (TN influent)<sup>-1</sup>. A couple of studies also report no N<sub>2</sub>O emissions from wastewater reactors or from the whole wastewater treatment line (Ahn et al. (2010) and Wicht and Beier (1995) in Table 3). The literature shows that N<sub>2</sub>O EFs referring to reject water treatment tend to be in the upper part of the N<sub>2</sub>O EFs measured at wastewater reactors and on the wastewater line. Conversely, N<sub>2</sub>O EFs referring to sludge treatment are in the lower part of the N<sub>2</sub>O EFs measured at wastewater reactors and on the wastewater line. Whole plant N<sub>2</sub>O EFs are only reported by Yoshida et al. (2014b), using the MTDM, and are in the upper part of the literature range (Table 3).

Although fewer studies normalised N<sub>2</sub>O emissions according to nitrogen removal (as kg N<sub>2</sub>O-N (TN removed)<sup>-1</sup>), Table 4 reports an N<sub>2</sub>O EF pattern similar to that described in Table 3. In Table 4, N<sub>2</sub>O EFs vary between 0.006 and 2.8% (as kg N<sub>2</sub>O-N (TN removed)<sup>-1</sup>), while EFs based on measurements taken for reject water treatment tend to be at the higher end of the literature range.

The international guideline provided by the Intergovernmental Panel on Climate Change for quantifying N<sub>2</sub>O emissions from WWTPs states that N<sub>2</sub>O emissions might only be important when facilities are equipped with process units performing nitrification and denitrification steps for nitrogen removal (IPCC, 2006). However, the same guideline seems to contradict this statement when it suggests the use of the EF reported by Czepiel et al. (1995) (Table 3), which was obtained measuring N<sub>2</sub>O emissions from a WWTP not performing nitrogen removal. Table 3 and Table 4 show that WWTPs equally emit N<sub>2</sub>O, thereby disregarding their ability to remove nitrogen. Nevertheless, the literature reports only three studies carried out at plants without nitrogen removal, while 22 studies refer to measurements at facilities capable in this regard (Table 3 and Table 4).

In general, the literature reports a very large range of CH<sub>4</sub> and N<sub>2</sub>O EFs, suggesting that emissions are driven by plant-specific conditions.

**Table 3.** Literature review of N<sub>2</sub>O emission factors (EFs) expressed in percentage as kg N<sub>2</sub>O-N (kg TN influent)<sup>-1</sup>.

Source	Method	Nitrogen removal	Value (min – max)	Study
Reactors	OSPM	NA	0.001	Sümer et al. (1995)
Reactors	OSPM	Yes	0.001 – 0.040	Benckiser et al. (1996)
Reactors	OSPM	Yes	0.01 – 0.08	Kimochi et al. (1998)
Reactors	OSPM	NA	0.02	Sommer et al. (1998)
Reactors	OSPM	Yes	2.5	Kampschreur et al. (2008)
Reactors	OSPM	Yes	0.02 – 0.08	Joss et al. (2009)
Reactors	OSPM	No	0.00 – 1.65	Ahn et al. (2010)
Reactors	OSPM	Yes	0.006 – 1.546	Ahn et al. (2010)
Reactors	OSPM	Yes	3.2 – 5.7	Desloover et al. (2011)
Reactors	OSPM	Yes	0.04	Aboobakar et al. (2013a)
Reactors	OSPM	Yes	2.8	Daelman et al. (2013)
Reactors	OSPM	Yes	0.09 – 0.12	Ren et al. (2013)
Reactors	OSPM	Yes	1.25 – 4.15	Sun et al. (2013)
Reactors	OSPM	Yes	0.12	Rodriguez-Caballero et al. (2014)
Reactors	OSPM	Yes	0.013 – 0.197	Brotto et al. (2015)
Reactors	OSPM	Yes	0.37 – 1.24	Masuda et al. (2015)
Reactors	OSPM	Yes	0.007 – 2.170	Schaubroeck et al. (2015)
Reactors	NA	NA	0.318	Thomsen (2016)
Wastewater line	OSPM	No	0.014 – 0.055	Czepiel et al. (1995)
Wastewater line	OSPM	NA	0.0 – 9.3	Wicht and Beier (1995)
Reject water treatment	OSPM	Yes	1.46	Kampschreur et al. (2008)
Reject water treatment	OSPM	Yes	0.62	Gustavsson and la Cour Jansen (2011)
Reject water treatment	OSPM	Yes	1.0 – 1.3	Schaubroeck et al. (2015)
Sludge treatment	OSPM	NA	0.013 – 0.070	Oshita et al. (2014)
Whole plant	MTDM	Yes	0.10 – 2.72	Yoshida et al. (2014b)

Values are reported either as single or as interval. Reactors refer to the process unit in Figure 1, named “Wastewater reactor”. OSPM: On-site point measurements. MTDM: Mobile tracer gas dispersion method. NA: Not available.

**Table 4.** Literature review of N<sub>2</sub>O emission factors (EFs) expressed in percentage as kg N<sub>2</sub>O-N (kg TN removed)<sup>-1</sup>.

Source	Method	Nitrogen removal	Value (min – max)	Study
Reactors	OSPM	Yes	0.03 – 0.11	Kimochi et al. (1998)
Reactors	OSPM	No	0.006 – 3.055	Ahn et al. (2010)
Reactors	OSPM	Yes	0.006 – 2.800	Ahn et al. (2010)
Reactors	OSPM	No	0.24	Townsend-Small et al. (2011)
Reactors	OSPM	Yes	0.76	Townsend-Small et al. (2011)
Reactors	OSPM	Yes	0.11 – 0.15	Ren et al. (2013)
Reactors	OSPM	Yes	0.03 – 0.56	Yan et al. (2014)
Reactors	OSPM	Yes	0.50 – 1.67	Masuda et al. (2015)
Reject water treatment	OSPM	Yes	0.25 – 1.08	Joss et al. (2009)
Reject water treatment	OSPM	Yes	0.13 – 0.57	Christensson et al. (2013)

Values are reported either as single or as interval. NA: Not available. Reactors refer to the process unit in Figure 1, named “Wastewater reactor”. OSPM: on-site point measurements.

## 2.4 Current limitations of carbon footprint assessments of wastewater treatment plants

The carbon footprint assessment of a WWTP is a plant environmental indicator that provides the overall contribution of the facility to climate change. Such an assessment is a useful tool for GHG mitigation actions, because it helps find the most effective mitigation strategies identifying the factors causing the highest impacts on climate change, and because it can verify the effects of mitigation actions already fulfilled. Additionally, the evaluation of the WWTP carbon footprint helps in understanding the contribution of these facilities to the global emission of anthropogenic GHGs. The carbon footprint assessment accounts for fugitive CH<sub>4</sub> and N<sub>2</sub>O emissions, while it neglects fugitive emissions of CO<sub>2</sub>, due to their biogenic origin (IPCC, 2006).

The literature reports only two studies accounting for the annual contribution of a WWTP to climate change (Gustavsson and Tumlin, 2013; Yoshida et al., 2014a), therefore assessing the carbon footprint for accounting purposes. Gustavsson and Tumlin (2013) studied 16 Scandinavian WWTPs while adopting an inconsistent accounting method and usually using data that did not refer to the investigated facility, e.g. fugitive N<sub>2</sub>O EF referring to a study in Australia. Conversely, Yoshida et al. (2014a) assessed the carbon footprint of only one WWTP, using plant-specific data and the life cycle assessment (LCA) as consistent accounting method.

The LCA is a standardised method (ISO, 2006a; 2006b), which evaluates environmental impacts related to the use of resources and emissions into the environment from all processes involved in the study. Usually, an LCA is carried out by analysing several impact categories (e.g. eutrophication, human toxicity, etc.), but it can also focus on one category only. The evaluation of impacts on climate change impact category is called a “carbon footprint” (JRC, 2011). An LCA can be used for two main purposes, depending on the modelling framework used. The attributional modelling framework is used for accounting purposes, namely accounting for environmental impacts within the inventory time period, e.g. one year (JRC, 2011). Conversely, the consequential modelling framework is used for supporting decisions from alternative options (JRC, 2011). The two modelling frameworks are not comparable, though, because they involve dissimilar inventory data collections, impact assessment result calculations and LCA result interpretations (JRC, 2011).



A literature review by Zang et al. (2015) revealed that the LCA using the consequential modelling framework was largely applied in wastewater treatment investigations, and it showed the importance of fugitive GHG emissions from WWTPs when evaluating the carbon footprint. When fugitive CH<sub>4</sub> and N<sub>2</sub>O emissions are not included in the assessment, some studies have shown that the most important driving factor in carbon footprint evaluation is energy consumption (Niero et al., 2014; Rodriguez-Garcia et al, 2011). Additionally, Gustavsson and Tumlin (2013) and Yoshida et al. (2014a) reported the importance of fugitive GHG emissions, but only the latter used plant-specific GHG measurements. Nowadays, the carbon footprint of Danish and Swedish wastewater utilities is estimated using GHG EFs, which are based either on models or on measurements at plants located in different countries (DANVA, 2012; SVU, 2014). It is still unknown how much the use of fugitive GHG EFs, which are not plant-specific, leads to correct carbon footprint evaluation.

Misleading results can be obtained in the carbon footprint evaluation of a WWTP when certain relevant factors and processes are excluded from the assessment or data are not site-specific (Corominas et al., 2013; Yoshida et al., 2014a). The literature is still lacking a study investigating the importance of fugitive GHG emissions when plant-specific data are used for assessing the carbon footprint of several WWTPs that use different treatment stages and technologies, and when the assessment includes all steps involved in material handling, from its arrival at the plant to the final discharge into the environment.

## 3 Methodology

This chapter reports the methods and tools used for meeting the PhD thesis objectives. The Scandinavian facilities studied herein are described initially, while the following two sections explain the methods used to measure fugitive gas emissions. Another section provides a description of national and international guidelines commonly used in South Scandinavia for fugitive GHG emission estimations and reporting from WWTPs. Finally, a description of carbon footprint evaluation using the LCA method is provided.

### 3.1 Facilities studied

Seven WWTPs were chosen to represent the technologies and process units usually used at conventional facilities in South Scandinavian. The WWTPs were very different from each other: Table 5 and Table 6 describe the wastewater and sewage sludge lines of the plants, while Table 7 reports the use of energy and additional processed material.

Besides WWTPs, a biogas plant in Linköping (Sweden) was also studied. The plant treated organic waste mainly consisting of household food waste (48%), food industry waste (28%) and slaughterhouse waste (20%). Waste was pre-treated in an enclosed hall aerated by a blower that led the exhaust air to a biofilter for odour emission treatment. In a homogenisation tank, the waste was mixed and subsequently kept for one hour at a temperature of 70°C. The substrate was fed to four anaerobic digesters working with wet fermentation, as in WWTPs. The produced digestate was stored in an open tank and eventually used on agricultural land. Generated biogas was temporarily stored in a gasholder and subsequently treated with activated carbons for hydrogen sulphide removal. Chemical scrubbers, and occasionally water scrubbers, were used for upgrading the biogas, which was finally compressed and sold as vehicle fuel.

**Table 5.** Wastewater treatment lines of the studied wastewater treatment plants.

<b>Plant (country)</b>	<b>Population equivalent (x 1000)</b>	<b>Wastewater Reactors</b>	<b>Reject water treatment</b>	<b>Tertiary treatment</b>
Lynetten (DK)	750	Bio Denipho	None	None
Avedøre (DK)	265	Bio Denitro	None	None
Lundtofte (DK)	150	Bio Denitro and Anaerobic MBBR	None	Membrane bioreactors
Holbæk (DK)	60	Sequencing batch reactors	Deammonification process	Sand filter
Ryaverket (SE)	805	Activated sludge reactors, Nitrifying trickling filters, Denitrifying moving bed biofilm reactors	None	Disc filters
Källby (SE)	120	Activated sludge reactors	None	Flocculation and clarification ponds
Växjö (SE)	95	Activated sludge reactors, HYBAS reactors	Deammonification process	Sand filter

HYBAS: combination of activated sludge reactors and moving bed bioreactors.  
The population equivalent describes the treatment capacity of the plant.

**Table 6.** Sewage sludge treatment lines of the studied wastewater treatment plants.

<b>Plant (country)</b>	<b>Mixed sludge storage and treatment</b>	<b>Digestate storage</b>	<b>Digestate treatment</b>	<b>Biosolids treatment and fate</b>
Lynetten (DK)	Outdoor in enclosed tanks	Outdoor in enclosed tanks	Indoor thickening and dewatering	On-site incineration
Avedøre (DK)	Indoor	Outdoor in open tanks	Indoor thickening and dewatering	On-site incineration
Lundtofte (DK)	Outdoor in enclosed tanks	Indoor	Indoor thickening and dewatering	On-site incineration
Holbæk (DK)	Outdoor in open tanks	Outdoor in open tanks	Enclosed thickening and dewatering	Outdoor stockpiles for 2 days (UoL)
Ryaverket (SE)	Outdoor in enclosed tanks	Indoor	Indoor thickening and dewatering	Outdoor stockpiles for 3 weeks (UoL)
Källby (SE)	Outdoor in open tanks	Outdoor in open tanks	Outdoor thickening, indoor dewatering	Enclosed silos for 1 day (UoL)
Växjö (SE)	Indoor	Indoor	Indoor thickening and dewatering	Outdoor stockpiles for 26 weeks (UoL)

UoL: Use on agricultural land. Since land application occurs every 26 weeks, biosolids produced in Holbæk, Ryaverket and Källby are stored in a different facility before being used on land.

**Table 7.** Energy use and additional processed material in the studied wastewater treatment plants.

Plant (country)	Biogas treatment	Energy output and use	Additional processed material
Lynetten (DK)	None	Methane to natural gas grid, Heat to district heating grid	Sewage sludge
Avedøre (DK)	Burnt in a combined heat and power engine	Electricity to the grid	Sewage sludge
Lundtofte (DK)	None	Methane to natural gas grid	None
Holbæk (DK)	Burnt in a combined heat and power engine	Electricity to the grid	Landfill leachate
Ryaverket (SE)	None	Methane as vehicle fuel	Sewage sludge, Food waste
Källby (SE)	Upgraded with chemical scrubbers	Methane as vehicle fuel; Heat to district heating grid	Sewage sludge
Växjö (SE)	Upgraded with chemical scrubbers	Methane as vehicle fuel	Sewage sludge, Food waste

## 3.2 Methods for emission quantifications

This section describes in detail the mobile tracer gas dispersion method and provides an essential description of the other measurement methods adopted by teams that collaborated on the PhD project.

### 3.2.1 Plant-integrated emission measurement approaches

The MTDM combines the known release of tracer gas with measurements of downwind atmospheric gas concentrations. Furthermore, the MTDM uses the properties of gases with long atmospheric lifetimes to maintain a constant concentration ratio during mixing and transportation in the atmosphere (Lamb et al., 1995; Stiversten, 1983). Therefore, when the tracer gas is continuously released at a constant rate from the emitting area, the real-time emission rate of the target gas can be calculated by comparing the detected plume traverse concentrations of the target and tracer gases, as shown in Eq. 1.

$$E_{tg} = Q_{tr} \cdot \frac{\int_{plume\ start}^{plume\ end} (C_{tg} - C_{tg\ baseline}) dx}{\int_{plume\ start}^{plume\ end} (C_{tr} - C_{tr\ baseline}) dx} \cdot \frac{MW_{tg}}{MW_{tr}} \quad (\text{Eq. 1})$$

where  $E_{tg}$  is the target gas emission in mass per time;  $Q_{tr}$  is the known tracer release in mass per time;  $C_{tg}$  and  $C_{tr}$  are the detected plume concentrations within the plume traverse in parts per billion (ppb);  $C_{tg\ baseline}$  and  $C_{tr\ baseline}$  are baseline concentrations of the target and the tracer gas (ppb), as shown in

Figure 2, and  $MW_{tg}$  and  $MW_{tr}$  are the molecular weights of the target gas and tracer gas, respectively (Scheutz et al., 2011). The integration of concentrations across the plume's cross-section minimise the influence of potential improper gas mixing and emitting source simulation with the target gas (Mønster et al., 2014). Acetylene ( $C_2H_2$ ) was usually used as a tracer gas, due to minimal possible interfering sources and a long atmospheric lifetime (23 days) (Logan et al., 1981).

Gas concentration measurements were carried out with a vehicle equipped with two gas analysers (having high detection frequency and good precision), a global positioning system device and a weather station. Gas analysers were based on cavity ring down spectroscopy. One gas analyser detected  $CH_4$  and  $C_2H_2$  twice per second (G2203, Picarro, Inc., Santa Clara, CA), with a precision of 0.77 and 0.06 ppb, respectively. The other gas analyser detected  $N_2O$  and  $C_2H_2$  every three seconds (S/N JADS2001, Picarro, Inc., Santa Clara, CA), with a precision of 7.7 and 0.6 ppb, respectively. Gas analyser precision is the reproducibility of a measurement and is defined as standard deviation when detecting a constant gas concentration for one hour. Atmospheric gas samples were taken from the roof of the vehicle and analysed, and detected concentrations were shown in real time on screens. Gas cylinders with calibrated flow meters were used to control the flow rate of released  $C_2H_2$ .

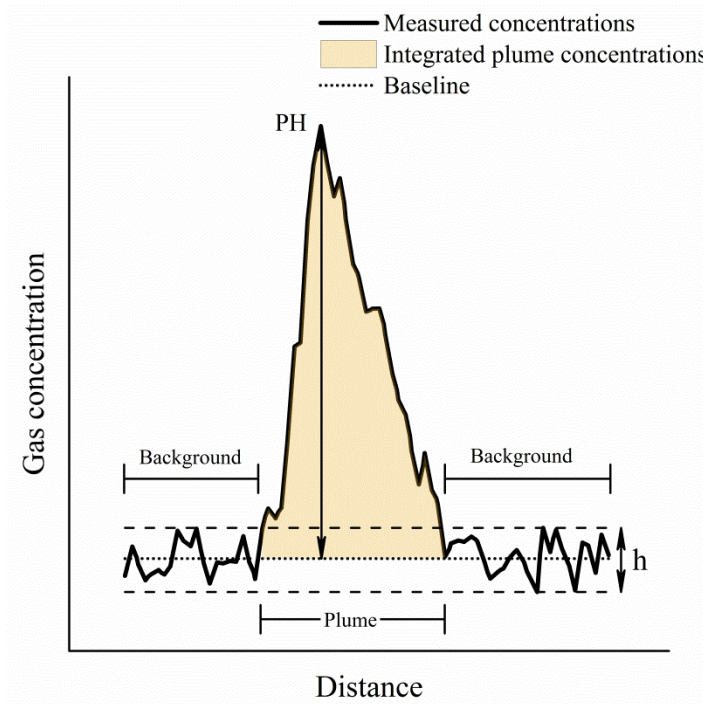


Figure 2. Example of a plume traverse (Paper I).

All measurement campaigns consisted of a screening phase (off-site and on-site) and a quantification phase. During the screening phase, the vehicle was driven around to measure atmospheric concentrations of target and tracer gases. Emitting sources were identified through a comparison of downwind and upwind concentrations of the studied gases. The screening phase started outside the facility, to ensure the absence of interfering off-site sources. Subsequently, screening inside the facility allowed the identification of on-site emitting sources. The main phases of a measurement campaign are shown in Figure 3. The on-site screening (Figure 3a) allowed the correct placement of the tracer gas cylinders (Figure 3b), to best simulate the target gas emission pattern. During the quantification phase, tracer gas was continuously released (Figure 3b) while the plume was traversed several times at a proper distance away from the emitting source measuring target and tracer gas (Figure 3c). This measuring distance away from the facility assured sufficient target and tracer gas mixing and allowed a suitable signal-to-noise ratio (SNR) in the gas concentrations recorded in each plume traverse. The signal-to-noise ratio is defined as suggested by Shrivastava and Gupta (2011):

$$SNR = \frac{PH}{0.5 h} \quad (\text{Eq. 2})$$

where PH is the maximum concentration above baseline measured in the plume traverse, and h is the difference between the highest and the lowest concentrations in the background (Figure 2) (Shrivastava and Gupta, 2011). A plume traverse was considered complete when the background concentration was identified on both sides of the plume (Figure 2).

At the biogas plant in Linköping (SE), the IDMM was applied (Paper IV). This method is based on the backward Lagrangian stochastic model used to quantify plant-integrated CH<sub>4</sub> emissions. The model input data were atmospheric CH<sub>4</sub> concentrations, and weather information averaged over 15 minutes. Atmospheric CH<sub>4</sub> concentrations were measured with an open-path gas analyser that was placed 100–200 m away from the biogas plant and 1.5 m high from the ground. Weather conditions were recorded using a weather station with a 3D sonic anemometer. For each time interval, the model simulated the route of 50,000 air parcels going backwards from the CH<sub>4</sub> concentration measurement path to the emitting area. Therefore, based on CH<sub>4</sub> concentration measurements and weather conditions, the model provided the CH<sub>4</sub> emission rates for each time interval (Paper IV).

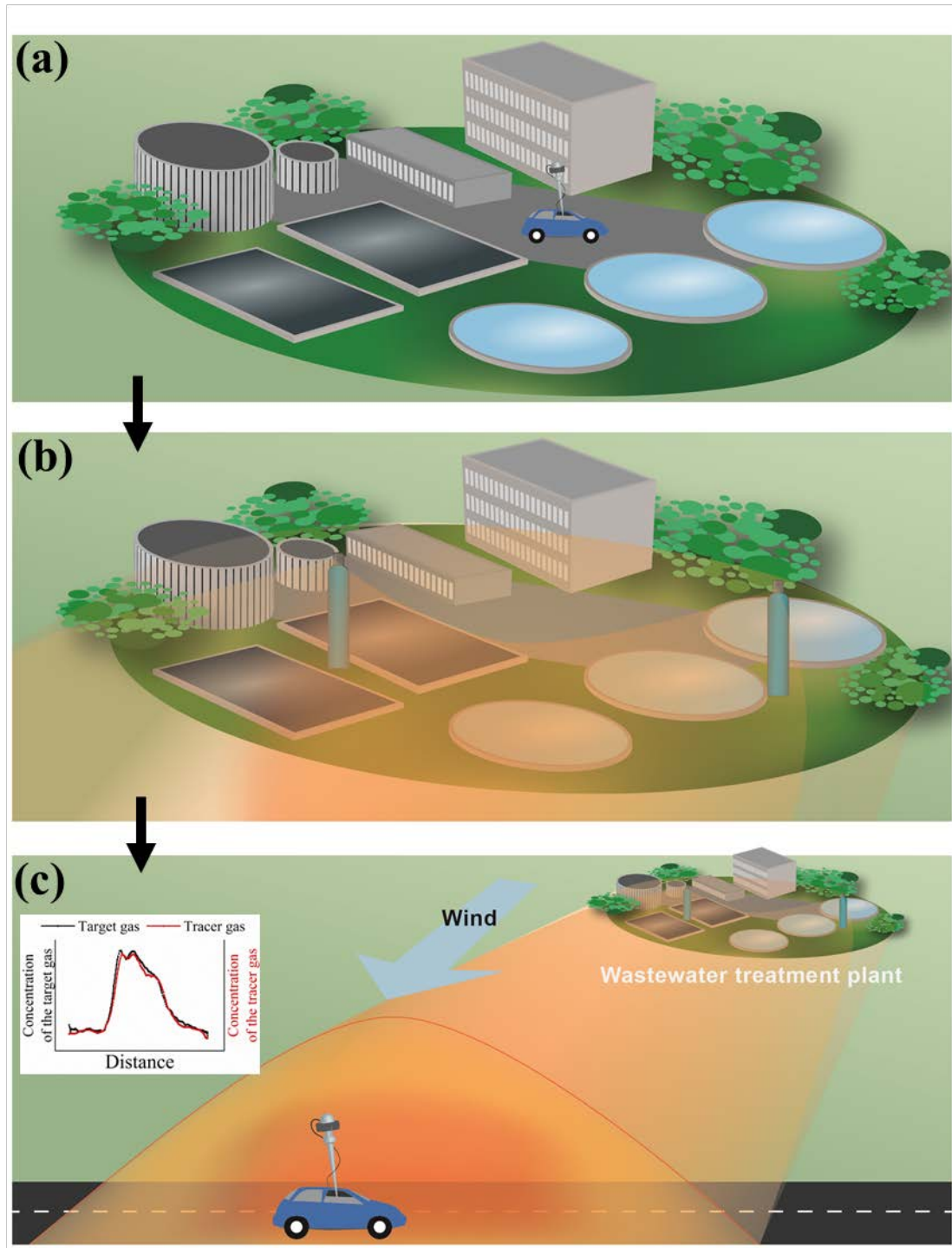


Figure 3. Application of the mobile tracer gas dispersion method at a wastewater treatment plant (Paper II). (a) The on-site screening phase. (b) Tracer gas placement and controlled release. (c) The quantification phase, when the plume is traversed for measurement of target and tracer gas concentrations.

At Ryaverket WWTP, plant-integrated ammonia ( $\text{NH}_3$ ) emissions were quantified using the solar occultation flux method (Paper III). This method is applied with a gas analyser, using the infrared radiation of the sun to determine the absorption properties of the target gas (Johansson et al. 2014; Mellqvist et al. 2010, 1999; Mikel and Merrill, 2011). Emission rates from the WWTP were obtained by driving the gas analyser across the plume so that vertically integrated gas concentration columns were measured using solar light that cut through the plume. An initial atmospheric background spectrum outside the source plume was used as a reference for consecutive plume spectra. The integrated mass of  $\text{NH}_3$  across the plume was obtained by summing consecutive gas column records. The  $\text{NH}_3$  emission rate was calculated by multiplying the integrated plume mass of  $\text{NH}_3$  by the average wind velocity of the plume.

### 3.2.2 On-site emission measurement approaches

At Lynetten and Ryaverket WWTPs, the MTDM was also applied to quantify emissions from specific process units (Paper II and Paper III), which was possible only at these two facilities, due to plant layouts.

At Ryaverket WWTP, on-site measurements were also performed by using two other methods: the static tracer gas dispersion method and the inferred flux method, both based on the same MTDM principle (Paper III). The static tracer gas dispersion method (STDM) used a static instrument, as previously applied for  $\text{CH}_4$  emission quantifications from leachate wells at landfills (Fredenslund et al., 2010). At Ryaverket WWTP, the STDM was applied to quantify emissions from the ventilated duct in the building where sludge thickening and digestate dewatering occurred.  $\text{C}_2\text{H}_2$  was released in the enclosed ventilated duct upstream of a fan, which allowed for the proper mixing of tracer and target gases where gas sampling occurred. The gas samples were taken at the end of the duct, where the gas analyser was placed at a fixed position. Emission rates of the target gases were obtained by using a variation of Eq. 1, whereby, instead of plume integration, just the ratio of the two gases was considered.

The inferred flux method was applied at Ryaverket WWTP when one of the target gases had a very low emission rate. The emission rate of the less abundant target gas was inferred from the multiplication of two factors: the emission rate of the more abundant target gas and the concentration ratio of the less abundant target gas to the more abundant target gas above background. The concentration ratio was obtained by analysing atmospheric samples at different points close to the source, to assure the proper homogeneous mixing



of the gases. The emission rate of the more abundant target gas was calculated with the MTDM.

At the biogas plant in Linköping, several on-site measurement methods were applied at different emitting sources by four different teams (Paper IV). CH<sub>4</sub> leakages from biogas-bearing plant components were initially identified while screening the area with infrared cameras, portable open-path CH<sub>4</sub> laser instruments and portable CH<sub>4</sub> concentration analysers. Later, leakages were quantified by using the high-volume sampling technique and the dynamic chamber method. The high-volume sampling technique was based on a high amount of air pumped through a hood encapsulating the CH<sub>4</sub> leakage. CH<sub>4</sub> concentrations were continuously recorded with a gas analyser, while the volume flow was measured with a differential pressure sensor and a calibrated orifice. The emission rate was calculated using Eq. (3):

$$E = V * \rho * (c_{out} - c_{in}) \quad (\text{Eq. 3})$$

where  $E$  is the emission mass flow (mg CH<sub>4</sub> h<sup>-1</sup>),  $V$  is the volume of the air flow ( $m_{Air}^3 h^{-1}$  STP, dry),  $\rho$  is the gas density of methane (mg ml<sup>-1</sup>) and  $c_{out}$  and  $c_{in}$  are the exhaust and background CH<sub>4</sub> concentrations (ppmv, corresponds to  $ml_{CH_4} m_{Air}^{-3}$ ), respectively.

The dynamic chamber method used a similar approach to that of the high-volume sampling technique, albeit with three main differences. First, less fresh air was pumped through the chamber, second, vane anemometers were used for measuring airflow and third, airflow in input and output was discontinuously sampled using evacuated glass vials and subsequently analysed in the laboratory, using a gas chromatography.

CH<sub>4</sub> emissions from the biofilter were quantified by following a method based on discontinuous air sampling operations under a foil covering the biofilter (Cuhls et al, (2016); Daniel-Gromke et al., 2015). The airflow volume was measured in the inlet pipe conveying air to the biofilter. Additionally, CH<sub>4</sub> emissions from the biofilter were quantified, disregarding the potential CH<sub>4</sub> oxidation action of the biofilter and applying standardised methods for gas flow quantifications from the inlet pipe of the biofilter (EN ISO 25140:2010 (2010) and EN 15259:2007 (2007)).

Standardised methods were also applied when measuring CH<sub>4</sub> emissions emanating from the exhaust pipe of the biogas upgrading unit (EN ISO 25140:2010 (2010) and EN ISO 25139:2011-08 (2011)). In this case, gas flow was obtained from the biogas plant's operational data.

CH<sub>4</sub> emissions from open tank digestate storage were quantified, using the static and dynamic floating chambers. CH<sub>4</sub> emission rates measured using the chambers (about 0.3 and 0.5 m<sup>2</sup>) were extrapolated to the entire surface of the open tank (about 1075 m<sup>2</sup>). When applying the floating static chamber method, gas samples inside the chamber were taken discontinuously at defined time intervals over 30 minutes. Emissions from the surface covered by the chamber (mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup>) were obtained by multiplying the volume-to-area ratio of the chamber (m<sup>3</sup> m<sup>-2</sup>) and the linear slope of the built-up CH<sub>4</sub> concentrations measured inside the chamber (mg CH<sub>4</sub> m<sup>-3</sup> h<sup>-1</sup>). The floating dynamic chamber method is the same method previously described as the “dynamic chamber method”, albeit using a chamber with a fixed volume floating on the emitting source.

### 3.3 Guidelines for greenhouse gas emission estimations

For comparison with the measured emissions, the IPCC and the Danish national guidelines were used in this study to estimate fugitive CH<sub>4</sub> and N<sub>2</sub>O emissions from the studied WWTPs.

For annual CH<sub>4</sub> emission estimation, the IPCC guideline suggests a tier-based approach (IPCC, 2006) via Tiers 1 to 3. The choice of tier depends on the data available. When no country-specific EFs are available, Tier 1 should be applied, using default values provided in the given model. Tier 2 is applied when country-specific EFs are available, by substituting the default EFs used in the Tier 1 model. Finally, when a country-specific method is available, Tier 3 should be used. In this study, Tiers 1 and 3 were used. Tier 1 models the biomethane potential of a specific facility, using a default CH<sub>4</sub>-producing capacity factor, a country-specific organic pollution load and the plant treatment capacity (population equivalent). Annual CH<sub>4</sub> emissions are then calculated by subtracting plant-specific CH<sub>4</sub> production and recalcitrant organic matter remaining in the biosolids from the biomethane potential. Within Tier 3, the Danish national guideline models annual CH<sub>4</sub> emissions from two main sources: the sewage system plus the wastewater line, by considering the pollution load, and anaerobic digestion, by considering plant-specific CH<sub>4</sub> production (Thomsen, 2016).

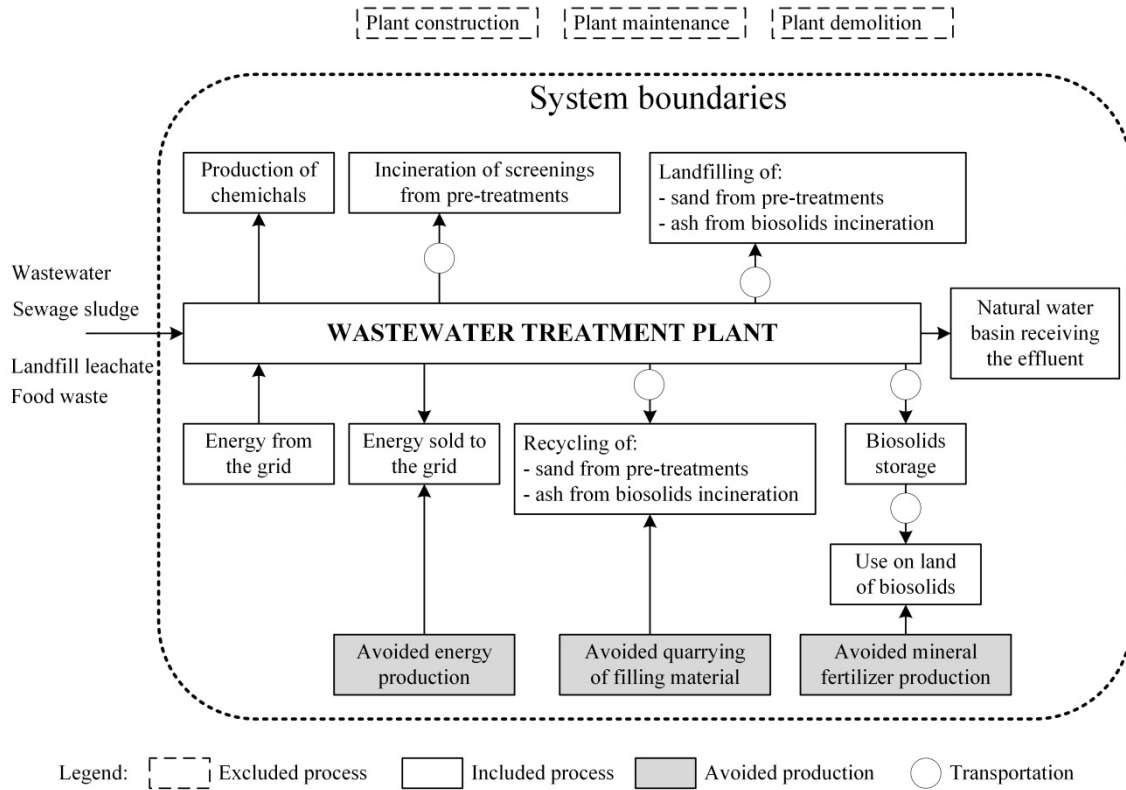
For annual N<sub>2</sub>O emission estimation, the IPCC guideline uses a default EF based on a plant-specific pollution load, which is expressed as “plant treat-

ment capacity” (population equivalent (PE)) ( $3.2 \text{ g N}_2\text{O PE}^{-1}$ , Czepiel et al. (1995)). Conversely, the Danish national guideline uses a country-specific EF ( $0.318\%$  as  $\text{kg N}_2\text{O-N (kg TN influent)}^{-1}$ ) multiplied by the annual WWTP nitrogen load (Thomsen, 2016).

### 3.4 Carbon footprint evaluation, using the life cycle assessment

The carbon footprint evaluation of the investigated WWTPs was carried out by following the iterative phases of the LCA (Paper V) (ISO, 2006a; 2006b), namely goal and scope definition, life cycle inventory, life cycle impact assessment and results interpretation. European Commission guidelines were also followed (JRC, 2010; 2011). The goal of the study was to evaluate the carbon footprint of seven WWTPs in 2015. The functional unit was set to 1 Mg of input material with a site-specific composition. The boundaries of the carbon footprint evaluation included all stages of material treatment, from its arrival at the facility to its release into the environment (Figure 4). Since the evaluation focused only on the operational phase in 2015, plant construction, maintenance and demolition were neglected in the assessment (Figure 4).

Primary and secondary data were gathered in the life cycle inventory phase. Primary data were plant-specific and consisted of the consumption of energy taken from the grid, the amount of energy delivered to the grid, the consumption of fuels and chemicals, the composition of input and output material and fugitive GHG emissions. Primary data were based on environmental reports on wastewater utilities, communications with plant operators and plant-integrated measurements (Paper II, Paper III, Yoshida et al., 2014b). Secondary data, which included the production of energy, fuels and chemicals, were chosen according to their technological, geographical and temporal relevance and taken from internationally recognised databases.



**Figure 4.** System boundaries of the carbon footprint evaluation (Paper V). Not all reported processes were significant for all investigated wastewater treatment plants.

The life cycle impact assessment only analysed climate change impact category, including CH<sub>4</sub>, N<sub>2</sub>O and fossil CO<sub>2</sub> emissions (IPCC, 2013). Sensitivity analysis and uncertainty propagation were used to investigate the results of the life cycle impact assessment phase (Clavreul et al., 2012). With the contribution analysis, the main contributors to the carbon footprint were identified, whereas with the perturbation analysis, the most sensitive parameters in the model were found. Very sensitive parameters were those causing a large change in the carbon footprint evaluation when a small change occurred in their value.

The effect of using different approaches for estimating fugitive GHG emissions was investigated through a scenario analysis comparing plant-specific measurements, Danish national guidelines and IPCC guidelines (Paper II; Paper III; IPCC, 2006; Thomsen, 2016). Moreover, a scenario analysis compared the results of the carbon footprint, using CH<sub>4</sub> EFs measured during normal operating conditions and digester malfunctioning for the Avedøre and Väjjö WWTPs (Paper II; Yoshida et al., 2014b).

The uncertainty of the carbon footprint was quantified by running uncertainty propagation. For each plant, the most sensitive parameters were run with their probability distributions in 100,000 Monte Carlo simulations.

Site-specific WWTP models (Tables 5–7) were built in EASETECH v2.3.6 (Clavreul et al., 2014), which is a mass flow-based LCA tool. Energy and mass balance was carried out for all WWTPs, all of which were modelled as “one process”. The use of district heating and electricity available to the grid was an input accounted for as a carbon footprint burden, due to emissions related to the production of the energy. On the contrary, methane, heat and electricity produced inside the WWTP, but sold and used outside, were outputs accounted for as carbon footprint savings, due to the avoided production of conventional energy available in the grid. The carbon footprint evaluation did not account for energy produced and consumed by the WWTP, because this energy was neither an input nor an output of the WWTP. The use of fuel was considered as an energy input into the plant. Carbon footprint savings were obtained when the production of new material was avoided by using the WWTP output material (Figure 4).

All WWTPs were modelled with specific input material compositions and four different material outputs. The effluent conveyed to the natural water recipient was the first output. Sand removed in the pre-treatments was the second output. The third material output was made by screenings removed in the pre-treatments and sent to an external incinerator, modelled in EASETECH (Møller et al., 2013). The fourth material output was either biosolids or ash from the on-site incineration of biosolids. Off-site storage of fresh biosolids accounted for CH<sub>4</sub> and N<sub>2</sub>O emissions according to specific measurements (Paper III). The model representing the use of biosolids on agricultural land was based on EFs reported by Bruun et al. (2016), and on mineral fertiliser substitution ratios suggested by Danish ministry guidelines (Ministry of Environment and Food of Denmark, 2015). Finally, material transportation was modelled in the software.

## 4 Results and discussion

### 4.1 Analysis of mobile tracer gas dispersion method

#### 4.1.1 Comparison among different instruments

For the first time, the MTDM was applied, using three analytical instruments that traversed the plume simultaneously and at the same distance away from the emitting source (Paper I). Analytical instruments had different detection frequencies (from 0.06 Hz to 2 Hz) and precisions<sup>2</sup> in measuring CH<sub>4</sub> and tracer gases (from 0.3 to 21.1 ppb, according to the gas). This plurality of CH<sub>4</sub> and tracer gases detections allowed the quantification of plant-integrated CH<sub>4</sub> emission rate from a WWTP using seven combinations of instrument and tracer gas. Since the instruments measured different tracer gases, the release precision ratio (RPR) was introduced to compare performances of the different instruments. RPR is the ratio between the molar release rate of the tracer gas and analytical instrument precision, and so it indicates the relative strength of the downwind signal for a given instrument measuring a specific gas. The measurement campaign consisted of two tracer gas-releasing tests, namely Test 1 and Test 2, in which 12 and 17 plume traverses were performed, respectively. The average CH<sub>4</sub> emission rate for each test was given as the average value of all plume traverses, and corresponding uncertainty was expressed as the standard error of the mean (SEM).

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<sup>2</sup> Precision indicates the consistency of repeated concentration measurements. In this study (Paper I), gas analyser precision was expressed in ppb and reported as three times the standard deviation of concentrations recorded over six minutes when a gas with a constant concentration went through the analytical instrument. The reported precisions refer to tests performed on-site on each analytical instrument when they were fed by a constant, but unknown, atmospheric gas concentration (Paper I).

The CH<sub>4</sub> emission rate quantified by each instrument and tracer gas combination was compared to the mean emission rate quantified by all instrument and tracer gas combinations. Table 8 reports that emission rates differed from the mean value by 1 to 15% in Test 1, and by 4 to 18% in Test 2. The CH<sub>4</sub> emission rate quantified by each instrument and tracer gas combination was also compared to the emission rate quantified by instrument and tracer gas combination 2, having the highest tracer gas RPR and measuring in both tests. Table 9 shows that instruments with a lower tracer gas RPR tended to provide greater emission rate uncertainty in Test 1, while they provided a lower emission rate in Test 2.

RPR is believed to be an important factor when applying the MTDM, because it influences the signal-to-noise ratio of the plume traverse measurements. A higher RPR provides plume traverses with a higher signal-to-noise ratio, which decreases the risk of choosing the wrong plume baseline concentration used in Eq. 1 (section 3.2.1).

**Table 8.** Comparison of different instrument and tracer gas combinations applying the mobile tracer gas dispersion method (Paper I). Differences in emission rate compared to the mean CH<sub>4</sub> emission rate quantified by all combinations.

Instrument and tracer gas combination	Difference in emission rate (%)				Tracer gas RPR	
	Test 1		Test 2		Test 1	Test 2
	Average	SEM	Average	SEM		
1	7	3	NA	NA	542.2	519.5
2	-1	4	11	2	153.8	153.8
3	-15	4	NA	NA	59.6	59.6
4	3	4	10	2	48.1	48.1
5	-12	4	-4	2	18.3	18.3
6	10	6	-18	2	18.0	16.0
7	9	4	NA	NA	18.0	17.2

NA: Not available. SEM: uncertainty expressed as standard error of the mean. Instrument and tracer gas combinations are ordered from the highest to the lowest tracer gas release precision ratio (RPR). Results are reported for two tracer gas-releasing tests.

**Table 9.** Comparison of different instrument and tracer gas combinations applying the mobile tracer gas dispersion method (Paper I). Differences in emission rate compared to the mean CH<sub>4</sub> emission rate quantified by instrument and tracer gas combination 2.

Instrument and tracer gas combination	Difference in emission rate (%)				Tracer gas RPR	
	Test 1		Test 2		Test 1	Test 2
	Average	SEM	Average	SEM		
1	8	3	NA	NA	542.2	519.5
2	0	4	0	2	153.8	153.8
3	-15	4	NA	NA	59.6	59.6
4	4	4	-1	2	48.1	48.1
5	-11	4	-13	2	18.3	18.3
6	11	6	-26	2	18.0	16.0
7	10	4	NA	NA	18.0	17.2

NA: Not available. SEM: uncertainty expressed as standard error of the mean. Instrument and tracer gas combinations are ordered from the highest to the lowest tracer gas release precision ratio (RPR). Results are reported for two tracer gas-releasing tests. Instrument 2 was chosen as a reference because it was the instrument with the highest tracer gas RPR measured in both tracer gas-releasing tests.



#### 4.1.2 Estimation of the method detection limit

When applying the MTDM, the lowest measurable emission is a function of three factors: the analytical instrument, weather conditions and the measurement distance. Therefore, the detection limit refers to specific conditions occurring in the measurement campaign. A novel approach to estimating the MTDM detection limit was developed using inverse Gaussian plume modeling (Paper II), in which the plume traverse is described with a Gaussian distribution. The plume's concentrations depend on the source emission rate, distance away from the source, emission height above ground level, surface roughness and atmospheric conditions. The model is described in Eq. (4):

$$C(x, y, z) = \frac{Q}{2\pi u \sigma_y(x) \sigma_z(x)} \cdot \left( e^{-0,5\left(\frac{z-H}{\sigma_z(x)}\right)^2} + e^{-0,5\left(\frac{z+H}{\sigma_z(x)}\right)^2} \right) e^{-0,5\left(\frac{y}{\sigma_y(x)}\right)^2} \quad (\text{Eq. 4})$$

where  $C$  is the concentration ( $\text{kg m}^{-3}$ ) in any given downwind plume point  $(x, y, z)$  measured from the source,  $Q$  is the emission rate ( $\text{kg s}^{-1}$ ),  $u$  is wind speed ( $\text{m s}^{-1}$ ),  $\sigma_y(x)$  and  $\sigma_z(x)$  are the dispersion coefficients (m) and  $H$  is emission height above ground level (m).

The lowest measurable emission rate  $Q_{min}$  was calculated from Eq. (4) by accounting for the parameters as follows. The smallest measurable downwind peak plume concentration  $C_{min}(x, 0, 2)$  was assumed as three times the magnitude of the background noise of the target gas determined for the specific measurement period (Shrivastava and Gupta, 2011).  $C_{min}(x, 0, 2)$  is the concentration in the middle of the plume traverse ( $y = 0$ ),  $x$  metres away from the source, and at 2 metres above the ground ( $z = 2$ ), where atmospheric concentrations were measured. Horizontal and vertical dispersion coefficients,  $\sigma_y(x)$  and  $\sigma_z(x)$ , were obtained following Briggs (1974) and according to atmospheric stability classes and plume transportation in open countryside or an urban area. Atmospheric stability classes were determined according to Pasquill (1974), using on-site wind speed measurements, insolation and cloud cover information.

#### 4.1.3 Assessment of uncertainty, due to emission height

At WWTPs, CH<sub>4</sub> emissions can occur from an elevated height, e.g. the top of a digester. If plume traverses are carried out too close to the plant, there is a risk of underestimating the CH<sub>4</sub> emission, because the tracer gas released from the ground does not mix completely with the target gas. A novel way to assess the potential underestimation of the target gas emission rate, due to elevated emission heights, was presented with an indicator called “Underestimation due to Tracer Height release” (UTH) (Paper II). UTH estimates in the emission rate quantification the potential bias caused by the vertical misplacement of the tracer gas, and it is expressed mathematically by Eq. 5:

$$UTH (\%) = \left| \left( 1 - \frac{PC_{tracer\_g}}{PC_{tracer\_h}} \right) \cdot 100 \right| \quad (\text{Eq. 5})$$

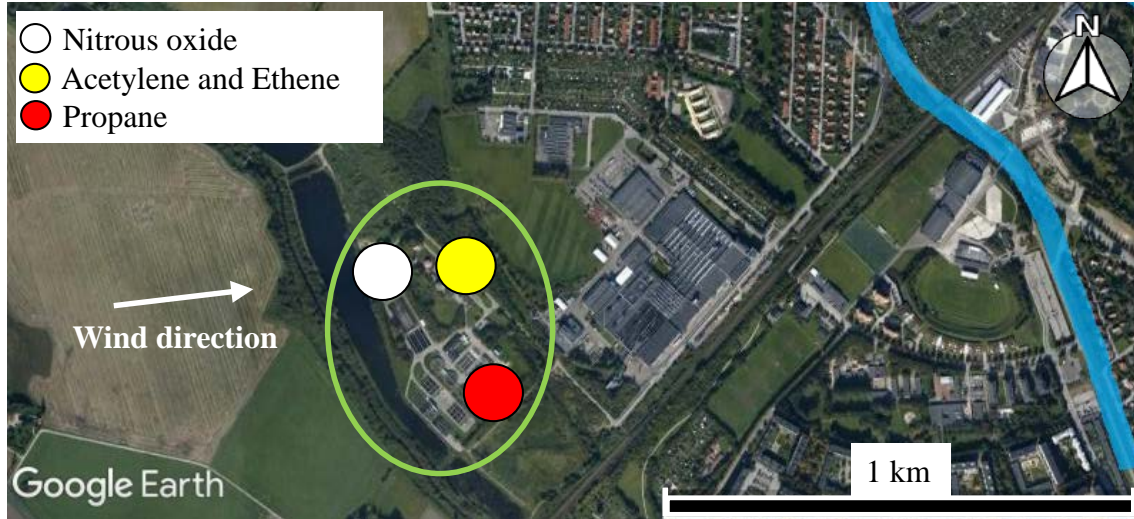
where  $PC_{tracer\_g}$  and  $PC_{tracer\_h}$  are the downwind peak concentrations (ppb) of the tracer gas estimated by the Gaussian plume model when the tracer is released from ground level and from an elevated height, respectively (Eq. 5). Therefore, UTH refers to specific measurement conditions, as  $PC_{tracer\_g}$  and  $PC_{tracer\_h}$  are based on Gaussian plume modelling, which uses the specific input parameters mentioned in section 4.1.2.

#### 4.1.4 Importance of tracer gas placement

A proper simulation of the emitting source with correct tracer gas placement was analysed by quantifying plant-integrated CH<sub>4</sub> emission rates from a WWTP, using four different tracer gases (Paper I). Figure 5 shows the tracer gas placements and the plume traverse position used for this analysis. Acetylene and ethene were placed correctly by the main on-site emitting source, while nitrous oxide was placed about 150 m upwind of the source, and propane was placed about 250 m sideward from the source (Figure 5). Even though wastewater treatment plants are known to emit nitrous oxide, emissions during this specific measurement campaign were below the MTDM detection limit (0.4 kg N<sub>2</sub>O h<sup>-1</sup>), corresponding only to 2.4% of nitrous oxide released as a tracer gas. The MTDM detection limit was calculated as reported in section 4.1.2. The same measurement platform performed 12 plume traverses, detecting CH<sub>4</sub> and the four tracer gases simultaneously.

The sideward misplacement only caused an overestimation of the emission rate of 12%, while upwind misplacement caused an overestimation of almost 50%, due to different distances travelled by the methane and the tracer gas

(Table 10). These findings confirmed previous studies (Mønster et al., 2014; Taylor et al., 2016), which used Eq. 1 (section 3.2.1) for emission rate quantification and reported higher errors when the tracer gas was misplaced upwind in comparison to sideward misplacement.



**Figure 5.** Tracer gas placement and plume traverse position when testing tracer gas misplacement (Paper I). The plume traverse position is marked with a blue line. The wastewater treatment plant is marked with a green circle.

**Table 10.** Tracer gas placement analysis (Paper I). Differences in emission rate compared to the mean  $\text{CH}_4$  emission rate quantified using acetylene and ethane as tracer gases released at the  $\text{CH}_4$  source.

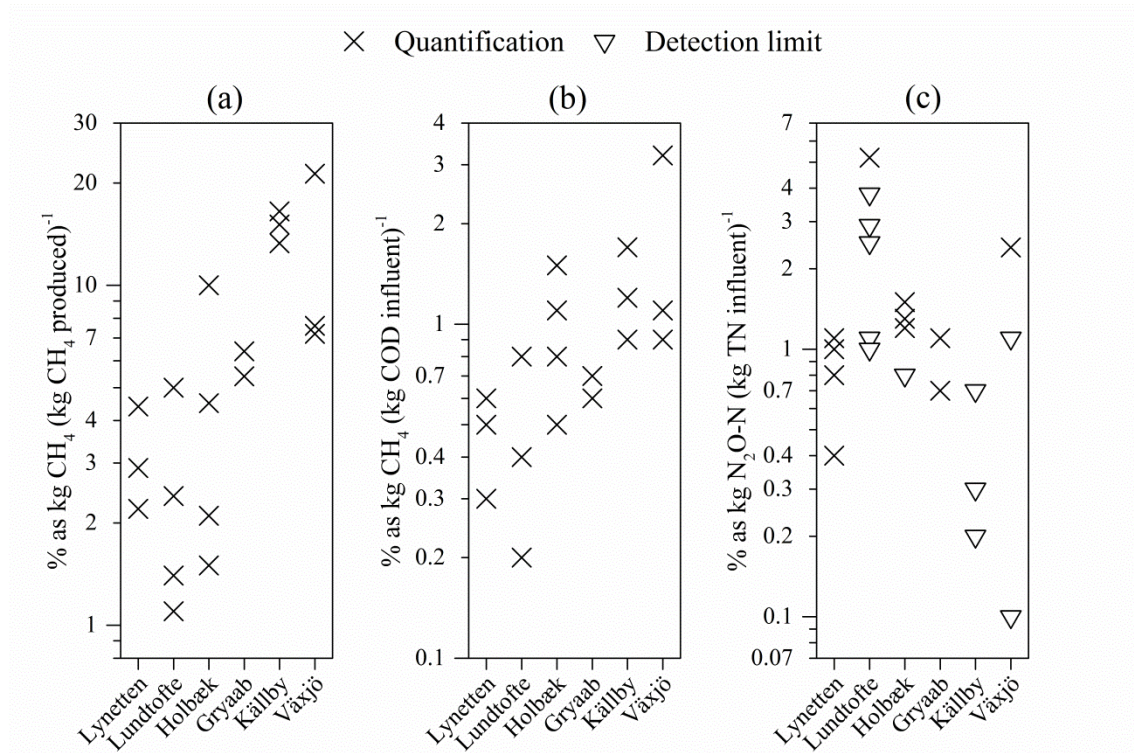
Tracer gas	Tracer gas placement	Difference in emission rate (%)	
		Average	SEM
Acetylene	Correct	9	3
Ethene	Correct	-9	3
Propane	Sideward misplacement	12	5
Nitrous oxide	Upwind misplacement	49	6

SEM: uncertainty expressed as standard error of the mean.

## 4.2 Quantification of plant-integrated emissions

When quantifying plant-integrated emissions from a facility, all smaller emitting sources from the large area are included and accounted for as one, to describe emissions from the whole facility. Therefore, plant-integrated measurements allow comprehensive emission quantification, which is very useful for emission reporting and the carbon footprint evaluation of the whole facility. Figure 6 shows plant-specific  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emission factors (EFs) based on plant-integrated measurements carried out using the MTDM at six WWTPs (Paper II and Paper III). Overall,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  EFs were plant-specific and in the upper range of EFs reported in the literature.

Plant-integrated  $\text{CH}_4$  emission rates were between 1.1 and 39.5 kg  $\text{CH}_4 \text{ h}^{-1}$ . Corresponding  $\text{CH}_4$  EFs were between 1.1% and 21.3% as kg  $\text{CH}_4$  (kg  $\text{CH}_4$  production) $^{-1}$  and between 0.2% and 3.2% as kg  $\text{CH}_4$  (kg COD influent) $^{-1}$  (Figure 6a and Figure 6b, respectively). Variations in EFs between different measurement campaigns were relatively small, with two exceptions. At Växjö, the highest  $\text{CH}_4$  EF referred to a measurement campaign when a leak at the top of one anaerobic digester let biogas escape from the reactor. Conversely, at Holbæk, the highest  $\text{CH}_4$  EF (Figure 6a) referred to a measurement campaign with an unusual 70% lower  $\text{CH}_4$  production compared to the other campaigns. The different  $\text{CH}_4$  EFs among the WWTPs could be explained by comparing sewage sludge and biogas treatments among the facilities. The lowest  $\text{CH}_4$  EFs were found at Lynetten and Lundtofte, which stored mixed sludge, digestate and biosolids in enclosed tanks and incinerated biosolids on-site (Table 6). In this way,  $\text{CH}_4$  emissions were avoided from the decomposition of organic matter contained in the stored material. A higher  $\text{CH}_4$  EF at Holbæk, in comparison to Lundtofte and Lynetten, could be explained by the open-air storage of fresh biosolids, digestate and mixed sludge (Table 6) and by the combustion of biogas in a combined heat and power engine (Table 7), which emits  $\text{CH}_4$  due to incomplete combustion. At Lynetten and Lundtofte, the produced biogas is delivered to the natural gas grid, without undergoing any treatment. At Källby and Växjö, open-air sewage sludge treatment (Table 6) and biogas upgrading units (Table 7) could be the reason for higher  $\text{CH}_4$  EFs compared to the other WWTPs (Figure 6).  $\text{CH}_4$  EFs at Ryaverket were the lowest among the Swedish WWTPs, perhaps due to sewage sludge treatment in enclosed units (Table 6) and the absence of biogas upgrading units at the plant (Table 7).



**Figure 6.** Plant-specific emission factors (EFs) based on plant-integrated measurements using the mobile tracer gas dispersion method in different measurement campaigns (Paper II and III). All EFs are expressed in percentage. (a) CH<sub>4</sub> EF as kg CH<sub>4</sub> emitted per kg CH<sub>4</sub> produced. (b) CH<sub>4</sub> EF as kg CH<sub>4</sub> emitted per kg COD influent. (c) N<sub>2</sub>O EF as kg N<sub>2</sub>O-N emitted per kg total nitrogen (TN) influent.

Plant-integrated N<sub>2</sub>O emission rates were between  $< 0.1$  and  $6.4 \text{ kg N}_2\text{O h}^{-1}$ , and corresponding N<sub>2</sub>O EFs were between  $< 0.1\%$  and  $5.2\%$  as kg N<sub>2</sub>O-N (kg TN influent)<sup>-1</sup>. In some measurement campaigns, N<sub>2</sub>O EFs were based on the MTDM detection limit (Figure 6c), which usually occurred when little N<sub>2</sub>O was emitted from a large emitting source like wastewater reactors. These emission characteristics involved a long measurement distance away from where instrument precision could not distinguish the target plume from the background concentration. Weather conditions were also important when N<sub>2</sub>O emissions were measured. Since unstable weather conditions cause a higher vertical dilution of gases than stable weather conditions, atmospheric instability worsens the distinction between plume and background concentrations.

Based on the numerous measurement campaigns carried out at facilities with different layouts, during different weather conditions and using different ana-

lytical instruments (Paper I-IV), a best practice MTDM application at WWTPs was formulated (Box 1).

At Ryaverket WWTP,  $\text{NH}_3$  plant-integrated emissions were also quantified (Paper III).  $\text{NH}_3$  emissions mainly occurred from the sewage sludge line, and the  $\text{NH}_3$  EF was equal to 0.09% as  $\text{kg NH}_3\text{-N (kg TN influent)}^{-1}$ . This is the first time that plant-integrated  $\text{NH}_3$  emission quantifications have been performed at a full-scale WWTP.  $\text{NH}_3$  emission quantifications are useful when carrying out environmental assessments, including impact categories focusing on the acidification and eutrophication of soil and water basins as well as the formation of particulate matter.

**Box 1. Best practice for the application of the mobile tracer gas dispersion method (MTDM) at wastewater treatment plants (WWTPs).**

The success of the MTDM application is a combination of several factors discussed below.

**Analytical instrument and choice of tracer gas**

The analytical instrument should measure target and tracer gases with good precision and high detection frequency as this provides a better plume definition and a faster measurement execution, thus smaller measurement uncertainties and lower method application costs (Paper I). A good instrument precision is particularly important when emissions are to be quantified from large area sources, as this requires that measurements are performed at a long distance from the facility in order to obtain fully mixed target and tracer gases at the measuring location, whereby dispersion results in low concentration differences in the plume compared to background concentrations within the same plume traverse. A precision of 3.8, 0.7 and 0.5 ppb when measuring methane, nitrous oxide and acetylene, respectively, was found sufficient for measuring at WWTPs (Papers I-III). These precision values are defined as three times the standard deviation of six minutes' constant concentration reading (Paper I).

Excluding environmental issues and price, any long-lived atmospheric gas can be used as tracer gas in MTDM application, as long as the tracer gas can be measured by the analytical instrument used (Paper I). The use of an analytical instrument with a poor tracer gas precision is possible by increasing the release rate of the tracer gas.



### **Preliminary study of the WWTP**

Before a measurement campaign, a desk study should be performed. The facility is investigated in terms of performed treatments, used process units and technologies, physical boundaries and internal layout, including heights of structures bearing the target gasses. The desk study should identify potential on-site  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emission sources, and the potential roads/paths for the on-site screening. Also the surroundings of the facility must be carefully investigated in order to identify possible driveable roads that can be used for plume traversing. Additionally, this investigation should locate possible interfering target and tracer gas sources (e.g. landfills, biogas plants, sugar factories, composting plants, farms, etc.) in the vicinity of the WWTP. Necessary information can be obtained with the support of the plant operator and the use of satellite images available for example on Google Maps, Zoom Earth or other similar internet applications. Preferable wind directions and possible measurement roads/paths should be marked on a map and used as guidance during the measurement campaign, as wind direction can change during the measurements forcing changes to the measurement plan.

### **Planning of the measurement campaign**

The measurement campaign should be carried out when the wind blows in a preferable direction identified during the preliminary study.

Additionally, stable weather conditions are preferred due to a lower vertical atmospheric dispersion of gases and a consequent better plume definition within a plume traverse compared to measurements performed during unstable weather conditions. Therefore, measurements during the night or with overcast weather, thus in absence of solar radiation, are recommended. Measurements during stable weather conditions are especially important when measuring  $\text{N}_2\text{O}$  emissions as the emissions mainly occur from the wastewater treatment reactors, which are sources emitting small fluxes from a large surface. This requires that measurements are performed at long distance from the facility, which could result in a plume concentration very close to the background concentration within the plume traverse. In this situation, measurements during little vertical atmospheric dispersion improve the distinction between plume and background concentrations within a plume traverse.

Only in one situation, measurements during unstable conditions could be favourable. This is when a high emission rate of the target gas, the measurement distance and the analytical instrument precision allow a clear distinction of the plume from the background concentrations within the plume trav-

erse. In this case, measurements during unstable weather conditions increases the target and tracer gas mixing, which could reduce potential error caused by tracer gas misplacement.

Measurements should be performed during weather conditions that allow continuous transport of air in the atmosphere. Wind speeds of 2-5 m s<sup>-1</sup> are preferable (Mønster et al., 2014). However, lower wind speeds (about 1 m s<sup>-1</sup>) could be sufficient if the tracer gas is placed close to the source and the plume is traversed at a distance that allows a proper mixing between tracer and target gases (Paper I and II). The MTDM is not affected by rain or snow.

### **Off-site and on-site screening**

During the screening phase, the vehicle carrying the analytical instrumentation is driven around off-site and on-site to measure atmospheric concentrations of target and tracer gases. Emitting sources are identified by comparing downwind and upwind concentrations of the detected gases. The screening phase starts outside the facility and ensures the absence of interfering off-site sources between the WWTP and the measurement road where the plume is planned to be traversed, as well as off-site sources located upwind of the WWTP. Subsequently, a screening inside the facility allows the identification of on-site emitting sources and a correct tracer gas placement for proper target gas emission simulation (Paper I). On-site screening should be performed using any available road/path inside and immediately around the WWTP.

During the whole measurement campaign, information about wind speed and direction, atmospheric pressure and temperature can be recorded using a weather station placed on-site. Such information is particularly useful when estimating the MTDM detection limit.

### **Tracer gas release**

Depending on the emitting area (area size, emission pattern and magnitude) and the distance to the quantification road, several tracer gas-releasing points can be set. The target gas-releasing rate should be known and constant over the quantification phase, thus controlled for example by calibrated flow meters. An additional control of the released tracer gas should be performed by continuously monitoring the weight loss of the tracer gas cylinders. If a continuous record is not possible, the weight of the cylinders should be measured before and after the tracer gas-releasing period. Previous measurements showed that when using acetylene as tracer gas, a total release of 0.2-1.1 kg C<sub>2</sub>H<sub>2</sub> h<sup>-1</sup> from one or two releasing points can be used (Paper II). These release rates were valid when the used instrument detected acetylene with a



precision of 0.5 ppb. The tracer gas should be released so it represents the emission pattern of the facility, i.e. releasing points should be placed at the most important on-site emitting sources.

### **Quantification phase**

The plume should always be traversed at a distance that, at the same time, allows a proper mixing of the target and tracer gases, and a proper plume distinction from the background concentration within the plume traverse. In particular, when quantifying CH<sub>4</sub> emissions from facilities with high structures bearing biogas (e.g. digesters), the plume should be traversed at a distance where the plume is fully mixed in both the horizontal and vertical plane.

If real-time downwind measurements show that the tracer and the target gas concentrations do not follow each other within the plume traverse, the tracer gas does not correctly simulate the emissions of the target gas. In this case, either the tracer gas-releasing points can be changed, or the plume could be traversed at further distance to improve the mixing of target and tracer gases. A plume traverse is considered successful if the target and the tracer gases follow each other when describing the plume, and the plume is completely crossed, thus the background concentration of target and tracer gases can be identified at both sides of the plume (Figure 2 and Figure 3c).

During the quantification phase, the number of the successful plume traverses should be as high as possible in order to record potential target gas emission variations over time. However, a minimum of 10 successful plume traverses should be collected (Mønster et al., 2014).

### **Data processing**

Only successful plume traverses are used for calculation of the target gas emission rate. For each plume traverse, the emission rate of the target gas is obtained by multiplying three factors, namely the tracer gas mass release rate, the downwind concentration ratio of the target and tracer gases and the ratio of the target and tracer gases' molecular weights. The ratio of the target and tracer gases is found by integrating the plume concentrations of each gas in the same plume traverse (Mønster et al., 2014). Only the concentrations above plume baseline are included in the integration and ratio calculation. The plume baseline is found as the average of the concentrations on each side of the plume, namely the average of the concentrations measured in the background (Figure 2).

The target gas emission rate of one measurement campaign is reported as average emission rate calculated using all successful plume traverses, and

the standard error of the mean is used as estimate of the uncertainty. An estimation of the lowest detectable emission rate at a specific measurement campaign can be calculated by using inverse Gaussian plume modelling (section 4.1.2 and Paper II). Horizontal and vertical dispersion coefficients can be calculated following for example Briggs (1974) and according to atmospheric stability classes and plume transportation in open countryside or an urban area. Atmospheric stability classes can be established for example according to Pasquill (1974), using on-site wind speed measurements, insolation and cloud cover information. The estimation of the MTDM detection limit is crucial when little GHG is emitted from a large area, forcing measurements at a distance so far away that the analytical instrument, due to high atmospheric dilution, does not distinguish the plume from background concentrations within the plume traverse.

The indicator “Underestimation due to Tracer Height release” (UTH) should be calculated for all measurement distances used in calculating the target gas emission rate at a specific measurement campaign (section 4.1.3 and Paper II). The calculation of the UTH is only important when the plume is traversed relatively close to the WWTP with emissions occurring from elevated heights, because tracer gas released from the ground does not mix completely with CH<sub>4</sub> potentially emitted from the top of a CH<sub>4</sub>-bearing process unit. When the UTH value is higher than about 15%, the potential vertical tracer gas misplacement becomes significant if compared to other uncertainties in the CH<sub>4</sub> emission quantifications. However, target gas quantifications should always be performed at a distance resulting in a UTH of only few percent points.

### 4.3 Emission quantifications from on-site sources

Quantifying emissions from single on-site sources is of importance to plant operators when implementing emission abatement measures. These measures could have three objectives: decreasing the risks of explosions, since CH<sub>4</sub> is an explosive gas, optimising CH<sub>4</sub> recovery for higher profits with biogas sales and decreasing the environmental impacts of the facility.

When quantifying emissions from on-site sources, a preliminary screening phase is crucial to recognise all emitting sources. However, there is a risk that some sources will not be recognised, due to three main factors: the capabilities of the adopted instruments, the inaccessibility of some process units and, finally, variations in facilities’ operational states that influence emissions.

This risk was demonstrated when on-site CH<sub>4</sub> measurements were performed at the biogas plant in Linköping, where four different measuring teams, equipped with different instruments, did not identify the same on-site sources (Paper IV).

On-site CH<sub>4</sub> emission quantifications from the biogas plant showed that the main contributors were the digestate storage tank, pressure release valves at the compressor station and the water scrubber (Table 11) (Paper IV). Different measurement days showed different contributions to total emissions, due to fixing pressure release valve number 1 after on-site measurements on Tuesday, and water scrubber utilisation only on Wednesday. Results show that lower fugitive CH<sub>4</sub> emissions could be obtained by optimising digestate storage and avoiding the use of the water scrubber, which currently just supports the chemical scrubbers. Furthermore, better compressor station maintenance could increase plant profits through the sale of more CH<sub>4</sub>.

At Ryaverket WWTP, on-site CH<sub>4</sub> and N<sub>2</sub>O emission quantifications were performed over 13 measurement campaigns, scattered across eight months (from January to August 2015) (Paper III). To obtain an understanding of on-site emissions from different sources, process units were investigated multiple times. Table 12 reports the average CH<sub>4</sub> and N<sub>2</sub>O emissions quantified on site, showing that the main contributors to CH<sub>4</sub> emissions were the biosolid stockpiles, whereas the main contributors to N<sub>2</sub>O emissions were the nitrifying trickling filters. Therefore, fugitive GHG emissions from Ryaverket WWTP could be reduced by optimising the storage of biosolids as well as the wastewater nitrification process.

At Ryaverket WWTP, on-site NH<sub>3</sub> emissions were quantified at the same process units, as reported in Table 12 (Paper III). Results show that the best contributing on-site sources were the biosolid stockpiles and the building used for thickening and dewatering operations, accounting for 44 and 22% of total on-site NH<sub>3</sub> emissions, respectively. Therefore, NH<sub>3</sub> emissions from the WWTP could be reduced by optimising the highest emitting process units previously listed.

**Table 11.** Average on-site CH<sub>4</sub> emissions quantified at the Linköping biogas plant (Paper IV).

On-site CH <sub>4</sub> sources	Tuesday		Wednesday		Thursday	
	Emission rate (kg h <sup>-1</sup> )	Contribution to total (%)	Emission rate (kg h <sup>-1</sup> )	Contribution to total (%)	Emission rate (kg h <sup>-1</sup> )	Contribution to total (%)
Small sources	0.7	5	0.7	5	0.7	10
Pressure release valves (1 and 2)	7.6	54	0.3	2	0.3	5
Digestate storage tank	5.9	42	5.9	42	5.9	85
Water scrubber	NA	NA	7.07	51	NA	NA

For “contribution to total,” is meant total CH<sub>4</sub> emissions quantified on-site. NA: not available because the water scrubber was out of operation. At the compressor station, there were two pressure release valves emitting methane: number 1 emitted only on Tuesday, whereas number 2 emitted only on Wednesday and Thursday. Emissions from the digestate storage tank were only measured on Tuesday, but they were assumed constant during the three days.

**Table 12.** Average on-site CH<sub>4</sub> and N<sub>2</sub>O emissions quantified at the Ryaverket wastewater treatment plant (Paper III).

Process unit	CH <sub>4</sub> emission		N <sub>2</sub> O emission	
	Emission rate (kg h <sup>-1</sup> )	Contribution to total (%)	Emission rate (kg h <sup>-1</sup> )	Contribution to total (%)
Sand trap	1.8	9	0.01	< 1
Primary settlers	0.8	4	0.1	3
Activated sludge reactors	1.0	5	0.1	2
Nitrifying trickling filters	0.1	< 1	2.7	82
Post-denitrifying MBBR	0.01	< 1	0.1	2
Secondary settlers	0.4	2	0.2	5
Ventilation exhaust of thickening and dewatering building	2.3	11	0.02	1
Biosolid stockpiles	14.5	70	0.2	5

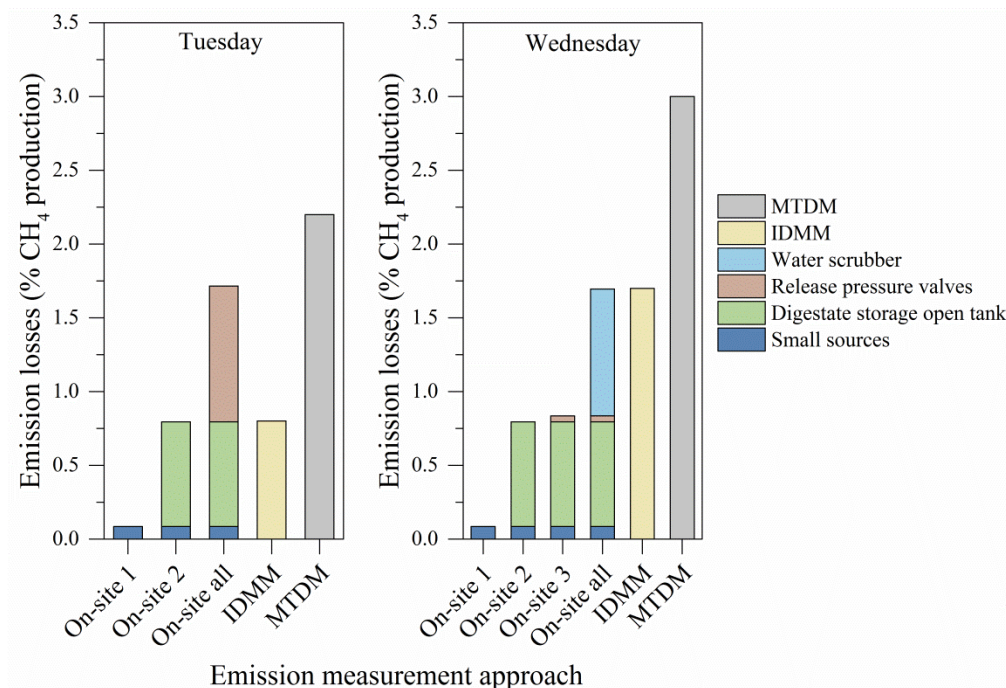
For “contribution to total” is meant total CH<sub>4</sub> or N<sub>2</sub>O emissions quantified on-site. A sand trap removes sand in preliminary treatments. Activated sludge reactors are wastewater reactors performing carbon removal. Nitrifying trickling filters are a wastewater technology performing wastewater nitrification. Post-denitrifying MBBRs are wastewater reactors performing wastewater denitrification, using moving bed bioreactors (MBBRs).

## 4.4 Comparison between plant-integrated and on-site measurement approaches

The application of plant-integrated and on-site measurement approaches was studied at the biogas in Linköping (Paper IV) and at Ryaverket WWTP (Paper III).

At the biogas plant in Linköping, the different measurement approaches were applied on the same days (Paper IV). Obtained CH<sub>4</sub> emission rates were influenced by the applied measurement method, the measuring equipment and the temporal emission variance, in general resulting in high and unknown uncertainties. For practical reasons, on-site CH<sub>4</sub> emitting sources (leakages or valves) were measured across different periods by different measuring teams. Due to weather and plant surroundings, plant-integrated emissions also were not measured simultaneously. Despite these differences, important findings were made, as summarised in Figure 7 and listed as follows:

- Using the on-site approach, very different plant CH<sub>4</sub> EFs were obtained if a large emitting source was not recognised during the initial screening, such as the water scrubber, release pressure valves or the digestate storage open tank (Figure 7).
- Plant CH<sub>4</sub> EFs were influenced by different emitting sources (pressure release valves and the water scrubber) on the two measurement days.
- On Wednesday, the inverse dispersion model method (IDMM) and on-site approaches obtained comparable CH<sub>4</sub> EFs. Conversely, on Tuesday, CH<sub>4</sub> EFs were different, most likely because IDMM was not applied when the pressure release valves were leaking.
- The MTDM provided larger EFs than other approaches because, besides measuring in different periods, it included emissions from other on-site sources. The open-air storage of food waste was not investigated by on-site measuring teams, but MTDM on-site screenings indicated emissions from this source. Additionally, emissions occurring from elevated heights at the facility would be included in the MTDM emission quantification but excluded by the on-site measuring teams and most likely by IDMM, because it measured relatively close to the plant due to gas analyser sensitivity.



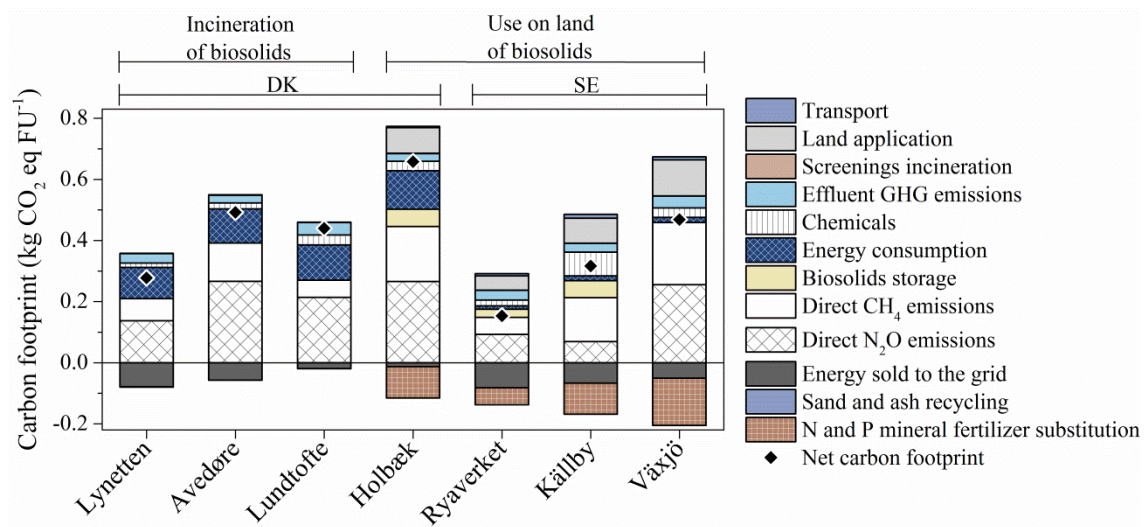
**Figure 7.** Average CH<sub>4</sub> emission factors obtained using different measurement approaches at the biogas plant in Linköping (Paper IV). Plant-integrated measurements were provided by the mobile tracer gas dispersion method (MTDM) and the inverse dispersion modelling method (IDMM). Both MTDM and IDMM were applied on the reported measurement days (Tuesday and Wednesday).

Also, at Ryaverket WWTP, on-site and plant-integrated measurements were performed (Paper III). In this case, CH<sub>4</sub> and N<sub>2</sub>O plant-integrated emission quantifications were about 1.5 times higher than emissions quantified on site. This difference could be explained by the combination of two factors, namely different measurement periods and potential on-site CH<sub>4</sub> sources not identified during screenings.

In general, plant-integrated measurements seemed to provide more comprehensive emission quantifications, whereas on-site approaches gave information about emissions occurring from specific sources. Therefore, plant-integrated measurements should be used for emission reporting and carbon footprint evaluation of the whole facility, whereas on-site approaches should be used to support emission mitigation strategies, which require the understanding of emissions from specific on-site sources.

## 4.5 Carbon footprint of the investigated wastewater treatment plants

None of the seven WWTPs could be considered carbon-neutral, due to their positive net carbon footprints, which ranged between 0.15 and 0.66 kg CO<sub>2</sub> equivalent per functional unit (Figure 8) (Paper V). Fugitive GHG emissions ranged between 44 and 71% of the total burden, resulting in the largest contributors to the carbon footprint. The sensitivity analysis showed that fugitive GHG emissions must be measured carefully, because they are very sensitive model parameters, and in this case, they were largely responsible for the uncertainty in the carbon footprint evaluation.



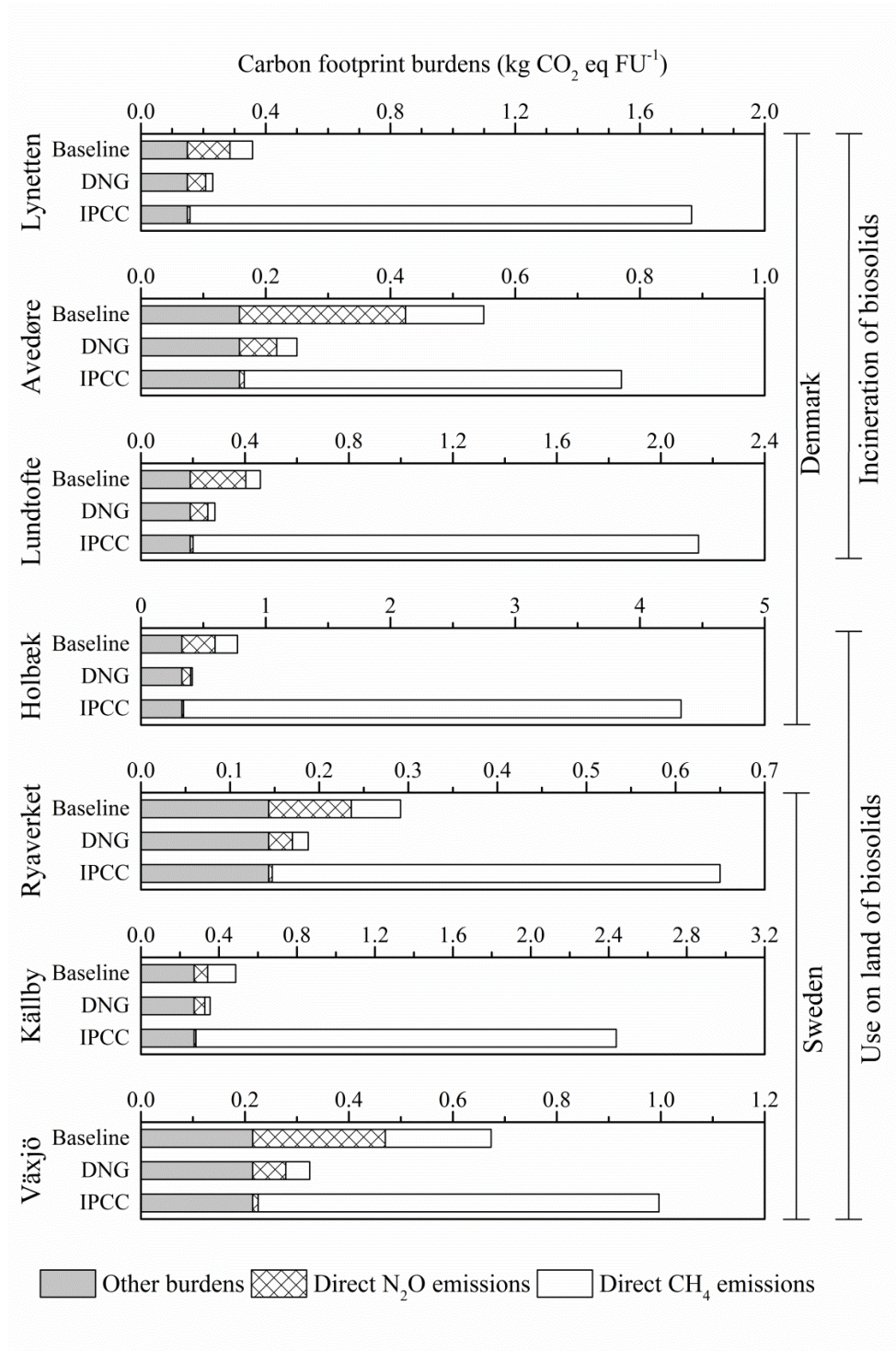
**Figure 8.** The contribution analysis of the carbon footprint for the studied wastewater treatment plants (Paper V). The functional unit (FU) was equal to 1 Mg of plant-specific inlet material in 2015.

The scenario analysis comparing different approaches for estimating the fugitive GHG emissions showed the importance of plant-specific measurements. At all investigated WWTPs, the approach used by the Danish national guideline (DNG) underestimated emissions of both CH<sub>4</sub> and N<sub>2</sub>O compared to the baseline approach using plant-specific measurements (Figure 9). Using the DNG led to an underestimation of the net carbon footprint between 1.7 times for Lundtofte and 3.9 times for Växjö. Even though the IPCC approach underestimated N<sub>2</sub>O emissions, its large overestimation of CH<sub>4</sub> emissions led to a GHG emissions overestimation compared to the baseline approach (Figure 9). Using the IPCC approach led to an overestimation of the net carbon footprint between 1.4 times for Avedøre and 7.2 times for Källby.

The scenario analysis comparing fugitive CH<sub>4</sub> emissions measured during normal operational conditions and digester malfunctioning showed a carbon footprint up to 320 times higher when digester problems occurred. This result suggests a careful monitoring strategy that accounts for emission variations in the inventory year.

Figure 8 shows that the contribution of energy consumption was very different for WWTPs operating in Denmark and Sweden. This difference was caused by the larger carbon footprint potential of the energy mix available in the Danish grid compared to the Swedish grid. In particular, the use of electricity for WWTPs operating in Denmark accounted for between 16 and 28% of the total carbon footprint burden. Conversely, for the Swedish WWTPs, the use of electricity caused only 2% of the total carbon footprint burden (Paper V). These very different impacts caused by electricity consumption occurred because electricity available in the Danish grid is mainly produced by wind, coal and natural gas, whereas electricity available in the Swedish grid is mainly produced by nuclear power and hydropower. The large difference in carbon footprint impacts due to energy consumption suggested the comparison of WWTPs, but only if operating within the same energy system.

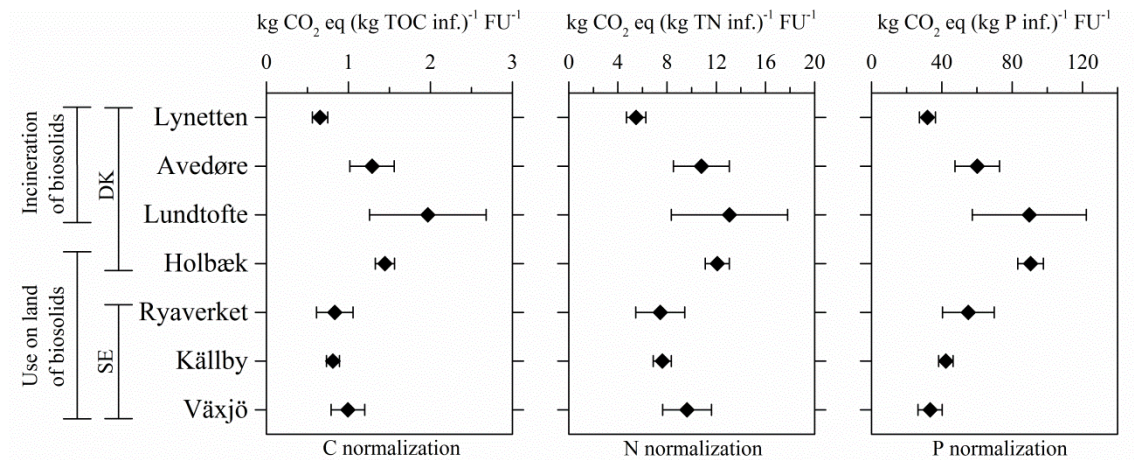




**Figure 9.** Scenario analysis of fugitive greenhouse gas emissions for the investigated wastewater treatment plants (Paper V). The carbon footprint burdens are expressed per functional unit (FU), equal to 1 Mg of site-specific input material in 2015. Three scenarios are shown: carbon footprint based on measured site-specific emissions (baseline), emissions estimated using Danish national guidelines (DNG) and emissions estimated using international guidelines (IPCC) (IPCC, 2006; Thomsen, 2016).

The impacts of different WWTPs could be compared by normalising net carbon footprints against pollution load (Figure 10). An additional support to this comparison was provided by the uncertainty propagation analysis performed in the EASETECH models, which allowed for calculating the net carbon footprint uncertainty for each plant.

Figure 10 shows net carbon footprints, normalised depending on different pollutants. WWTPs were grouped by country, due to the large influence that the energy system had on the results. Figure 10 shows a common pattern in ranking among the WWTPs. Lynetten and Lundtofte were the facilities with the smallest and the largest net carbon footprints, respectively, among the Danish WWTPs. Holbæk was the only Danish WWTP where biosolids were applied on agricultural land and reported one of the largest normalised net carbon footprints of the Danish facilities. The Swedish WWTPs showed comparable carbon footprints when outcomes were normalised by carbon and nitrogen loads. However, when results were normalised using phosphorus load, Källby and Våxjö reported a net carbon footprint that was lower than the value reported by Ryaverket.



**Figure 10.** Net carbon footprint of the studied wastewater treatment plants, normalised by the pollution load (Paper V). Error bars were obtained by propagating the uncertainty of the most sensitive model parameters with 100,000 Monte Carlo simulations. Results refer to the functional unit (FU), which is equal to 1 Mg of plant-specific inlet material in 2015.



## 5 Conclusions and future perspectives

### 5.1 Conclusions

The mobile tracer gas dispersion method (MTDM) was implemented and applied at six Scandinavian wastewater treatment plants (WWTPs) and at a Swedish biogas plant for plant-integrated methane ( $\text{CH}_4$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ) emission quantifications.

The MTDM was tested for plant-integrated  $\text{CH}_4$  emission quantifications, using simultaneously three different analytical instruments that measured  $\text{CH}_4$  and tracer gas concentrations with different detection frequencies and precisions (Paper I). Since analytical instruments could measure many tracer gases, plant-integrated  $\text{CH}_4$  emission rate was quantified using seven combinations of instrument and tracer gas. Emission rates differed between 1 and 18% from the mean emission rate quantified by all instrument and tracer gas combinations, showing fair agreement among the results. The strength of the downwind signal for a given instrument measuring a specific gas was defined by the release precision ratio (RPR), which is the ratio between the molar tracer gas release rate and analytical instrument precision. By using RPR, the plume detection of different instruments could be compared even if it was related to different tracer gases. In one tracer gas-releasing test, combinations with a lower tracer gas RPR tended to provide greater emission rate uncertainty, while in a second tracer gas-releasing test, combinations with a lower tracer gas RPR gave a lower emission rate. The RPR was identified as an important parameter in MTDM application, due to its influence on the signal-to-noise ratio of the plume traverses, which decreases the risk of choosing a wrong plume background concentration when calculating the emission rate of the target gas (Paper I).

A novel approach to estimating the MTDM detection limit was suggested by using the inverse Gaussian plume modelling (Paper II). The lowest measurable emission rate is a result of specific circumstances occurring during the measurement campaign, i.e. the analytical instrument's detection precision, weather conditions and the measurement distance away from the emission source. Estimation of the MTDM detection limit was found useful when little emission occurred from a large area, forcing measurements at a distance where the analytical instrument, due to high atmospheric dilution, could not distinguish the plume from background concentrations within the plume traverse.

A novel indicator, called “Underestimation due to Tracer Height release” (UTH), was suggested for assessing potential bias in emission rate quantification, due to vertical misplacement of the tracer gas (Paper II). Information provided by the UTH is useful when the plume is traversed relatively close to the plant. In this case, there is a risk of underestimating the CH<sub>4</sub> emission when the tracer gas released from the ground does not mix completely with CH<sub>4</sub> potentially emitted from the top of a CH<sub>4</sub>-bearing process unit (e.g. a digester). The calculation of the UTH in a measurement campaign assures that the potential vertical misplacement of the tracer gas has little influence (e.g. just a few percentage points) in the final emission rate quantification.

The importance of proper target gas simulation with correct tracer gas placement was shown by applying the MTDM for plant-integrated CH<sub>4</sub> quantification. A sideward misplacement of 250 m only caused an overestimation of the emission rate of 12%, while an upwind misplacement of 150 m caused an overestimation of almost 50%, due to different distances travelled by the methane and the tracer gas (Paper I).

CH<sub>4</sub> and N<sub>2</sub>O emission factors based on plant-integrated measurements using MTDM revealed that fugitive GHG emissions were plant-specific (Paper II and III). This result suggested different emission characteristics from facilities using different process treatments and technologies. Plant-integrated CH<sub>4</sub> emission rates were between 1.1 and 39.5 kg CH<sub>4</sub> h<sup>-1</sup>. Corresponding CH<sub>4</sub> EFs were between 1.1% and 21.3% as kg CH<sub>4</sub> (kg CH<sub>4</sub> production)<sup>-1</sup> and between 0.2% and 3.2% as kg CH<sub>4</sub> (kg COD influent)<sup>-1</sup>. Plant-integrated N<sub>2</sub>O emission rates were between < 0.1 and 6.4 kg N<sub>2</sub>O h<sup>-1</sup>, and corresponding N<sub>2</sub>O EFs were between < 0.1% and 5.2% as kg N<sub>2</sub>O-N (kg TN influent)<sup>-1</sup>.

At two facilities, plant-integrated GHG emissions were compared to emissions quantified using on-site measurement approaches (Papers III and IV). Plant-integrated measurements generally provided more comprehensive emission quantifications, which are useful for emission reporting and carbon footprint evaluation of the whole facility. Conversely, on-site approaches provided information about emissions occurring from specifically identified sources, which is important in the daily operation of the plant, on optimising treatment technologies and reducing emissions.

The carbon footprint evaluation of the studied WWTPs revealed that none of the facilities could be considered carbon-neutral, due to their positive net carbon footprints (Paper V). Fugitive GHG emissions were the largest contributors to the carbon footprint, causing between 44 and 71% of the total

burden, thereby showing that diffusive emissions are very important when assessing the impact of wastewater utilities on climate change. Additionally, fugitive GHG emissions were very sensitive model parameters and largely responsible for the uncertainty of the net carbon footprint evaluation. CH<sub>4</sub> and N<sub>2</sub>O emissions should be determined accurately, because a small change in their value produces a large change in the net carbon footprint.

The carbon footprint was evaluated by comparing different approaches to calculating fugitive GHG emissions. When default values from emission reporting guidelines were applied, the net carbon footprint was up to four times smaller or seven times larger compared to the measured plant-integrated GHG emission rates. This result shows the importance of plant-specific CH<sub>4</sub> and N<sub>2</sub>O measurements for properly quantifying anthropogenic GHG emissions caused by wastewater treatment.

Finally, the carbon footprint was evaluated by comparing fugitive CH<sub>4</sub> emissions measured during normal operating conditions and digester malfunctioning. The carbon footprint evaluation was up to 320 times higher when problems at digesters occurred, thereby suggesting a careful monitoring strategy accounting for emission variations in the inventory year.

## 5.2 Future perspectives

Despite the effort to fill the knowledge gap, the PhD project revealed also the need for further research into MTDM application and the carbon footprint evaluation of WWTPs:

- **Signal-to-noise ratio of the plume traverse:** The accuracy of emission rates obtained using the MTDM depends on the correct identification of the plume traverse baseline. Further research should identify the smallest value of the signal-to-noise ratio of the plume traverse that minimises the risk in choosing the wrong plume baseline.
- **Controlled release test:** the MTDM has been validated using a known release test from single point sources (Mønster et al., 2014), but a study about a controlled release test simulating diffusive emissions is still missing. Measurements at different distances away from the emitting source, and tracer gas placement, are two important factors to investigate in the controlled release test.
- **Future quantification of GHG emissions from WWTPs:** Since CH<sub>4</sub> and N<sub>2</sub>O emission factors were influenced by process units and treatments at the specific facility, the suggestion of new emission factors for national guidelines will not improve current GHG emission estimations at WWTPs. Instead, plant-specific measurements throughout the inventory year could provide more reliable results in this regard.
- **Measurement strategy and annual emission variations:** The measurement indicated large emission variations, even during normal operations, and during periods with operational disruptions (e.g. digester malfunctioning), the emission increased significantly. Future research should investigate the best measurement strategy throughout a year, in order to obtain representative annual GHG emission rates from a facility.
- **Harmonisation of measurement methods:** In this PhD project, several measurement methods were applied for GHG emission quantification, but only two of them were internationally standardised. Future research should focus on the harmonisation of measurement methods used for GHG emission quantifications at WWTPs. After the comparison study described in Paper IV, a harmonisation process for measurements at biogas plants was initiated by the project “MetHarmo – European harmonisation of methods to quantify methane emissions from biogas plants” (ERA-NET, 2017). The harmonisation of methods for measurements at WWTPs could use the re-

sults of the MetHarmo project, and implement further research especially for GHG emissions from the wastewater treatment line.

- **Carbon footprint evaluation as benchmark:** The carbon footprint could be used as an environmental benchmark for WWTPs, if systematic and transparent data collection supports consistent assessment modelling – as achieved in this PhD thesis. The current initiatives promoted by the Danish and the Swedish wastewater associations (DANVA, 2012; SVU, 2014) should be implemented following the specific directions provided in Paper V. For sound national benchmarking, wastewater associations should provide a consistent database containing secondary data that are useful for wastewater utilities within the same energy system.
- **Carbon footprint evaluation and storage of biosolids:** For the studied WWTPs, the storage of biosolids contributed up to 12% of the total carbon footprint burden (Paper V). GHG emissions from this process unit were based on measurements performed on biosolids no older than 3 weeks. Further research about CH<sub>4</sub> and N<sub>2</sub>O emissions from biosolid storage over six months, before land application, would allow for a more precise carbon footprint evaluation. The current literature provides information only about biosolids produced by sewage sludge that are not anaerobically digested (Larsen et al., 2017).





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## 7 Papers

- I. Delre A., Mønster J., Samuelsson J., Fredenslund A. M., Scheutz C., 2017. Emission quantification using the tracer gas dispersion method: the influence of instrument, tracer gas species and source simulation. Manuscript submitted to *Science of the Total Environment*, November 2017.
- II. Delre A., Mønster J., Scheutz C., 2017. Greenhouse gas emission quantification from wastewater treatment plants, using a tracer gas dispersion method. *Science of the Total Environment* 605–606, 258–268. doi:10.1016/j.scitotenv.2017.06.177
- III. Samuelsson J., Delre A., Tumlin S., Hadi S., Offerle B., Scheutz C., 2017. Optical technologies applied alongside on-site and remote approaches for climate gas emission quantification at a wastewater treatment plant. Manuscript submitted to *Water Research*, September 2017.
- IV. Reinelt T., Delre A., Westerkamp T., Holmgrend M. A., Liebetrau J., Scheutz C., 2017. Comparative use of different emission measurement approaches to determine methane emissions from a biogas plant. *Waste Management* 68, 173–185. doi:10.1016/j.wasman.2017.05.053
- V. Delre A., ten Hoeve M., Scheutz C., 2017. Site-specific carbon footprints of Scandinavian wastewater treatment plants using the life cycle assessment approach. Manuscript submitted to *Water Research*, November 2017.

In this online version of the thesis, **paper I-V** are not included but can be obtained from electronic article databases e.g. via *www.orbit.dtu.dk* or on request from.

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